

Dear Participant,

Thank for your interest in the PM Measurement Workshop to be held July 22-23, 1998 in Research Triangle Park, North Carolina. Enclosed you will find two background papers to help you prepare for your attendance. These papers are meant to complement one another and help us all make the most productive use of our short time together.

The first is a white paper prepared by EPA staff presenting the intended purpose of the Agency's PM "Super Sites" Program, and describing the relationship of this program to the rest of the US PM regulatory monitoring network. It was the anticipation of this program that gave rise to our workshop and lead to broader consideration of the many measurements needed to support PM research.

The second is a combined think piece prepared by panel of health, exposure, and atmospheric scientists offering their insights on the what, when, where, and how of ambient air research measurements needed to support review and implementation of national air standards and objectives in North America. This piece is meant to help us by focusing our thinking and giving us specific examples with which to work.

If I can be of any further assistance as you prepare for the PM Measurements Workshop please call Diana Phillips, at Personal Communications Service (919-544-4575), or me.

Dr. Russell W. Wiener
Chairman
Workshop Organizing Committee

Objectives of the PM “Supersites” Monitoring Program

Executive Summary

This document describes EPA’s rationale and underlying objectives for implementing a Supersites monitoring program, and provides background information prior to the July 22-23, workshop to be held in Chapel Hill, NC in 1998.

This program is being designed to conduct special, detailed chemical and physical characterization studies in geographic areas with a range of characteristic PM_{2.5} source-receptor and health risk situations. The scope and specific details of this program, termed “Supersites,” are being developed through substantial input from the scientific community, including the July 22-23rd workshop. The scope of this program includes the sampling and analysis of ambient aerosols and gases used to supplement particulate matter measurement, modeling, exposure and health risk assessment programs.

The EPA has not preselected geographic study areas, nor have we determined overall project design, the scope of measurements or sampling systems to be used. Development of the Supersites program is consistent with EPA’s desire to engage the scientific community in the design and operation of ambient air monitoring programs, and is responsive to recommendations in the National Academy of Sciences Report, *Research Priorities for Airborne Particulate Matter: I. Immediate Priorities and a Long-Range Research Portfolio*. As a focus for workshop discussions, several examples of research needs and associated measurements intended for coverage by this program are discussed below. These examples are meant to provide a common understanding of EPA’s objectives by a very diverse community of air quality professionals.

The geographic areas of the Supersites program (e.g., 4-7) ought to include regional variations in air pollution across the United States and include areas with unique characteristics (e.g., climatology, source distributions, air quality, population/demographics). For example, we know that differences exist in the composition and seasonality of aerosols in Southeastern, Northeastern, and Western U.S. cities. The number of Supersites will depend on the availability of sufficient resources to conduct high-quality, intensive and advanced measurement studies for ambient aerosols at each site commenced.

A single site viewed in isolation cannot address air quality issues that have strong regional components. Accordingly, these Supersites must be viewed as a complementary and intersecting activity to the regulatory monitoring network being deployed by the state and local agencies (Appendix C). Ideally, Supersites will be located where other major field studies (including exposure and health effects studies) either are in progress or being planned.

A major motivation for assembling the workshop is to bring together researchers from the atmospheric/physical science, health effects/exposure science, and regulatory communities to ensure that multiple objectives for studies in different disciplines can be addressed and coordinated across relevant programs. While the technical and scientific perspectives and objectives of these communities may differ, we suspect that major areas of common information needs exist. For example, epidemiological studies need to address co-pollutant interactions, which lead to the collection of the major components of particulate matter, as well as other gaseous species such as peroxides and ozone. Analogously, atmospheric processes underlying the formation of secondary aerosols are chemically coupled to oxidative species, including ozone and peroxides as examples. Ideally, this Supersites program will foster an environment across disciplines resulting in optimum use of environmental sampling resources. Accordingly, we acknowledge that the Supersites program is just one piece within an array of measurement and related studies.

There are three major Agency objectives common to all of the “Supersite” study areas:

- 1) support development of State Implementation Plans (SIP's) through improved understanding of source-receptor relationships leading to improved design, implementation, and tracking of control strategy effectiveness in the overall PM program;***
- 2) development of monitoring data and samples to support health and exposure studies to reduce uncertainty in National Ambient Air Quality Standards setting and to enable improved health risk assessments; and***
- 3) comparison and evaluation of emerging sampling methods with routine techniques to enable a smooth transition to advanced methods.***

The first two objectives should provide an opportunity to increase temporal, chemical, phase, and size fraction resolution of PM related measurements relative to “routine” monitoring programs that typically are limited, for example, to intermediate averaging times (e.g., 24 hours) and single size ranges. These knowledge gains will improve the scientific basis for setting standards and their implementation. The last objective recognizes the physical/chemical complexities of aerosols and associated sampling/analysis methodologies and the desire to accommodate and promote the use of emerging techniques.

Examples of the types of scientific and programmatic questions that will be supported through the Supersites program in coordination with other efforts are provided in Appendix A.

Program Objectives

Program objectives include three broad categories: 1) support for State Implementation Plans (SIP), 2) development of improved data for health and exposure studies and health risk assessments and 3) sampling method development and inter-comparisons. Since the Supersites program is an interdisciplinary effort bridging physical/chemical science and health assessment communities, a discussion of these objectives is provided to foster improved communications across these disciplines.

State Implementation Plan (SIP) Support

SIP support covers a very wide spectrum of activities that can be viewed as the atmospheric sciences component of this Supersites program. Activities that support both the development of effective emission control strategies and the continuous assessment of such strategies constitute SIP support. These activities range from developing qualitative insights regarding the nature and cause of a particular air pollution problem (e.g., regionally dispersed sulfate and carbon constitute the majority of PM_{2.5} aerosols), to a comprehensive application of complex three-dimensional gridded air quality simulation models. Included in the mix of SIP support tools are various source attribution/apportionment tools, air quality simulation models and observational approaches, and methods to characterize trends in air quality to track progress. These tools support planning of emissions control strategies and enable mid-course adjustments. A more detailed discussion of each of the following SIP activities, their role in SIP development, and their corresponding measurement needs is provided in Appendix B:

- Air Quality Characterization (beyond the routine chemical speciation program)
- Evaluation of Emission Estimates
- Air Quality Simulation Model Evaluation and Application
- Receptor and Observation-Based Models: Evaluation and Application of Advanced Methods
- Air Quality Trends and Tracking Progress of Control Programs

SIP activities use ambient measurements to drive and/or to evaluate the basic air quality management tools used to characterize and predict air quality in terms of temporal, spatial, size delineation and chemical composition coordinates. Each has a common objective of developing and tracking success of effective emissions strategies. The underlying SIP support tools include regional/urban scale air quality simulation models (e.g., MODELS 3) and a suite of more empirically-based observational methods. Air quality simulation models require emissions and meteorological input; whereas observational methods rely on ambient measurements to infer source to receptor relationships or preferred control strategy approaches. These latter methods include source apportionment techniques (e.g., CMB8, SAFER), and a group of methods that infer a generalized preferred precursor reduction approach without specifying source categories

(e.g., nitrogen oxides limit ozone formation more than volatile organic compounds at a particular location and time).

Supersites provide an important diagnostic complement to the routine monitoring program needed for air quality modeling and emissions inventory efforts. Supersites in a SIP context provide highly resolved measurements to diagnose the effectiveness of existing tools by uncovering their strengths and weaknesses. Supersites do not independently support SIP development as they clearly provide only a small fraction of the data needed for state implementation planning. The Supersites do provide operational support, e.g., direct input into source receptor models, and can be distinguished by recognizing their diagnostic support. Perhaps the largest area of technical criticism confronting the regulatory community over the last decade has been the lack of diagnostic measurements and techniques to support operational tools.

Exposure and Health Risk Assessments

Improved characterization of ambient particles and associated “toxic” constituents or co-pollutants are needed to address critical exposure and health effect issues. These issues include:

- Characterizing human exposure and the relationship of exposure to specific PM characteristics and related measurements collected at ambient monitoring sites,
- Identifying the causal agents and mechanisms for the acute and chronic health effects that are associated with PM_{2.5}.

Hypotheses regarding the potential causative agents, toxic mechanisms and potential for human exposures would be given consideration in selecting the chemical and physical methods to be deployed at the Supersites. For example, to evaluate the hypotheses that ultra fine particles, soluble metals, or electrophilic organic compounds are the causative agents, these species or properties would need to be measured. In addition, spatial and temporal factors will be considered to ensure that both acute and chronic effects and related exposures can be related to the ambient measurements at these sites and that sufficient temporal resolution and frequency of measurements will allow these hypotheses to be tested. Geographic considerations that require further study are the past observations suggesting that the health risks of PM_{2.5} are similar across diverse air sheds with different sources and background aerosols. To further explore this observation, it will be important to include a full range of geographic and source variations. Supersites, for example, may be augmented by additional neighborhood sites to assess the relationship among different exposure microenvironments of concern. Ideally, emerging information identifying and characterizing the nature of the “toxic” components of PM will be incorporated into the Supersite measurement program.

The Supersites, in addition to advancing the understanding of the chemical and physical

nature of PM and directly supplying ambient measures for exposure, epidemiologic, and clinical field studies, will potentially be an important resource for toxicology studies. For example, various types of samples of ambient particles could be collected for in-vitro and in-vivo toxicology studies addressing mechanistic questions.

Specific human exposure, dosimetry, toxicology, and epidemiology studies will be supported through resources available outside the Supersites program by EPA and other funding sources.

Monitoring Methods Development Platforms

Multiple and constantly changing demands are placed on the ambient air sampling and analysis community. There is a demand for information that provides greater resolution in chemistry, size distributions, and time which closely parallel new instrumentation developments. However, a transition (or collaboration with the expert community) is necessary before advanced methods can be used routinely by state and local agencies. As an example, increased time resolution of speciated aerosol measurements is often desired. State-of-the-science, continuous, in-situ, speciation samplers have now been developed. However, many of these methods require testing and comparison with standard methods to characterize the differences between techniques and to develop standard operating procedures. New methods must be evaluated before they are used at routine monitoring sites. The Supersites program represents an excellent opportunity to test new methods side-by-side with existing techniques and to allow for a smooth transition for the routine use of more advanced methods. Ideally, method comparisons should be performed in several different air sheds of varying characteristics to identify the weaknesses and strengths of various approaches. Clearly, these intercomparisons will identify several regional differences in performance between the Federal Reference Methods and techniques designed to capture all mass components. More exciting is the opportunity to accelerate deployment of continuous methods, improve organic carbon sampling and analysis techniques, and other methods providing particle-specific information. These sites should provide a vehicle for collaborations between the expert community (including universities, private R&D groups, industry, states & local agencies, and EPA) and State/local air monitoring organizations responsible for implementing "routine" monitoring platforms.

Relationships between Supersites and other components of the PM_{2.5} Monitoring Network

The Supersites complement the monitoring network in two very important ways: 1) as test platforms for application of advanced methods in routine networks, and 2) supply high resolution temporal, chemical, and size distribution data to enhance the less resolved data from routine sites. In turn, the routine networks complement the Supersites by providing strong spatial complements to the intensive Supersites. Appendix C provides an "Overview of the National

PM_{2.5} Monitoring Networks” including the routine chemical speciation program that will be enhanced by the Supersites.

Supersites will provide inter-comparison platforms for “routine” speciation samplers and Federal/Equivalent Reference Method mass samplers. Several issues related to the comparability of data from routine and advanced methods call for inter- and intra-method comparison. Supersites and routine sites need to be established and maintained to ensure progress in applying advanced methods routinely, and to reduce uncertainty in data trends interpretations brought about by changing methodologies.

The more resolved data from Supersites supports a wider spectrum of SIP and health risk assessment activities than provided by routine measurements alone. These include; air quality model evaluation, emissions evaluation, application of source-receptor methods, and support of health risk assessments.

Air quality is strongly influenced by multiple interacting spatial scales calling for characterization across super-regional, regional, urban, and local scales and various land use categories. A single site in an air shed is not capable of characterizing spatial gradients, background concentrations or transport phenomena that collectively interact to affect air quality at a specific location. Similarly, one site generally cannot reflect exposure everywhere in a large urban area with heterogeneous mix of sources. Consequently, a network of sites reflecting spatially disparate conditions is needed for most air quality assessments. Supersites should address both spacial and temporal characterization and may be complemented by the routine chemical speciation sites (e.g., which may serve as satellites). The Supersites could also provide vertical scale resolution (through optical techniques, elevated platforms, periodic aircraft flights) not expected to be part of routine networks.

In addition to routine monitoring programs, the Supersites design should take into account both existing and planned field studies conducted as by universities, industry and public-private organizations such as NARSTO. For example, the Supersites will likely focus on populated areas where health studies may also be conducted through other programs. Ideally, these “urban” sites should be coupled with intensive regional/background sites to delineate differences across different spatial regimes.

Appendix A: Questions Addressed Through Support of the Supersites Program

Examples of scientific and programmatic questions that will be supported through the Supersites program in coordination with other efforts are provided below:

Air Quality Management/Atmospheric Processes

What fraction of PM is locally, versus regionally, versus naturally (e.g., biogenic emissions) generated?

What are the various source categories contributing to PM and how much does each source category contribute to each of the above fractions?

What limiting conditions for the formation of PM_{2.5} exist now and over time with respect to coupled PM precursors (e.g., ammonia, nitrogen oxides, sulfur oxides, volatile organic compounds)?

How is the total PM, and the contributions of the various source categories, changing over time?

Are these changes in agreement with the expected changes based on increases or decreases in known, measured or estimated emission rates?

How well can regional scale and/or urban scale air quality models simulate the observed hourly, horizontal and vertical distribution of PM parameters and components?

Can a data base of hourly, horizontal, and vertical PM parameters and related components be used to improve air quality models? What are the spatial three-dimensional and temporal distributions of PM and oxidant precursors, PM components, PM and oxidant sinks and how are each transported at the surface, aloft, and in between?

How does the partitioning across gaseous and solid/liquid phases affect the fate of aerosol compounds?

What precursors (ammonia, nitrogen, and sulfur oxides, semi-volatile and volatile organic compounds) are most important to regional and local formation of secondary aerosols, and how is their relative importance expected to change over time?

How well can regional scale and/or urban scale air quality models simulate the observed

hourly, horizontal, and vertical distribution of PM parameters and components?

Can a data base of hourly, horizontal, and vertical PM parameters and components be used to improve air quality models?

What are the important chemical and physical coupling processes between oxidants and aerosols that affect the development of strategies targeted at both ozone and particulate matter? What regions of the country and over what seasons are coupling processes quantitatively important in developing co-pollutant emission strategies? How are these processes further coupled to deposition and toxics assessments?

What particle parameters, over and beyond those ordinarily measured, are useful in understanding sources? Examples include: particle number, particle surface area, particle size distribution, particle composition by size, light scattering, nonvolatile mass, nonvolatile plus semi-volatile mass, etc.

Exposure Assessment

Which parameters and components of PM are sufficiently evenly distributed across an urban area to provide a valid average community concentration (for use as a surrogate for the community average personal exposure)?

What exposure measure and time resolutions better predict the acute health effects than the 24-hr average? For example, is the maximum hourly concentrations or the 8-hour maximum concentration an independent exposure parameter or is it highly correlated with the 24-hour average?

What is the relationship between the ambient site measurements and human exposure?

Health Effects (Epidemiology/Toxicology)

What constituents of PM or associated pollutants are most highly associated with toxicity or adverse health effects?

What time resolution of ambient pollutant measures are the best predictors of adverse health effects (considering that this will vary with the type of health effect being studied and will be different between acute and chronic effects)?

To what extent does the presence of co-pollutants modify the effects of PM exposure?

Monitoring Methods

What is the difference in $PM_{2.5}$ that is characterized by Federal Reference Method techniques and techniques that capture a more comprehensive range of aerosol components?

How do such differences vary seasonally and geographically?

What is the difference in $PM_{2.5}$ that is characterized by Federal Reference Method techniques and techniques that capture a more comprehensive range of aerosol components?

How do such differences vary seasonally and geographically?

What operational and technical obstacles exist in applying emerging in-situ continuous methods for total $PM_{2.5}$ mass and spectate components, PM precursors, and oxidants/intermediates (peroxides, nitrogen dioxide, peroxy radicals, OH radical, nitrate radical)?

What steps must be deployed to move emerging methods into routine application?

Appendix B: SIP Support Activities and Data Needs

Examples of SIP support activities are provided here to illustrate how the Supersites program will support SIP's. The discussion is not intended to completely explain the technical approaches underlying the SIP process. Additional materials (Demerjian et al, 1995; EPA, 1996) provides a more thorough explanation of how the complementary uses of SIP support tools provide a solid underpinning of control programs. Throughout the discussion an emphasis is placed on how more detailed chemical, temporal and size distribution information from a Supersites will complement the routine chemical speciation program (Appendix C) which provides 24 hours averaged samples of major components (e.g., total organic carbon, not individual compounds) at less than daily sampling periods for one size cut (2.5 microns).

- **General Air Quality Characterizations.** Characterizing air quality through reconstructing component mass balances that identify the relative fractions of major components [e.g., major ions (sulfate, ammonium, nitrate), carbon (total elemental and organic), and elements (particularly crustal elements)] is an initial step leading to qualitative assumptions regarding principal impact sources and further refined analyses. This effort largely will be achieved through the routine chemical speciation program, which typically provides these measurements on 24-hour filter based samples. Since the routine program often will collect measurements on a 1/6 or 1/3 day sampling schedule, the Supersites program will enhance these general characterizations by providing daily (and probably continuous) measurements of the same components. This will allow for site-specific testing of the statistical adequacy of routine sampling schedules and allow for recommended adjustments to the routine program.
- **Evaluation of Emission Estimates.** Emissions are the major input into air quality simulation models and generally a source of considerable uncertainty. Emissions estimates include direct primary emissions, such as fugitives and soil/agricultural dust, incomplete combustion products, and condensible organic compounds; and precursors to secondarily formed aerosols, such as ammonia, nitrogen and sulfur oxides, and volatile organic compounds. While some of the precursor gases such as nitrogen and sulfur oxides are believed to be reasonably well characterized for major point sources, there remains large uncertainties in fugitives, ammonia and condensible organic compound sources. Many emission categories (and their near field concentration profiles) exhibit strong diurnal and seasonal patterns (e.g., mobile sources, biogenics), and emit specific organic compounds that are not analyzed routinely with standard chemical speciation protocols that sample over a 24-hour period. Similarly, assumptions regarding particle growth and related emissions size distributions (or particle number) incorporated in emissions models have large uncertainties that demand testing with observed data. Clearly, the

Supersites program will provide ambient data, which under certain conditions, will allow for evaluation (i.e., reality check) of modeled emissions estimates for certain species. Thus, the Supersites complements the routine networks by providing continuous (or at least higher time resolution) measurements and detailed species specific chemical information over various size fractions.

- **Application and Evaluation of Air Quality Simulation Models.** Emissions driven Air Quality Simulation Models (AQSM's) are valuable spatial, temporal, and chemical interpolators that require considerable validation with surface and aloft ambient measurements of both predicted values and process formulations. These modeling systems operate over multiple spatial scales (domains of over 1000 km and grid resolution down to 2-4 km, with capability for subgrid scale specific plume characterization), in near continuous time (effectively 1 hour increments) and considerable chemical definition. Given the enormous time, space, and chemical detail of modern AQSM's, there is no limit on the variety of ambient data that can provide useful testing information. While data from the routine programs (e.g., chemical speciation, PAMS) provide valuable evaluation data, those data fall short in either temporal (24 hours when hourly data are needed), chemical, or vertical resolution. Especially important is the availability of measurements that allow for true diagnostic¹ evaluation of the robustness of a model's ability to generate realistic emissions strategies by testing whether the model truly is capable of integrating transport and chemical process dynamics. In addition to common products (e.g., sulfate, nitrate, ozone) and precursor observations (nitrogen and sulfur oxides, volatile organic compounds); additional chemical data (that Supersites can provide) on precursors, such as ammonia; sinks (or removal species), such as peroxides and nitric acid (which also behaves as a precursor for aerosols), and important short lived and photochemical dynamic species, such as peroxy radicals and nitrogen dioxide allow for such testing. Due to the atmospheric chemical coupling between oxidants and aerosols, any evaluation of modern AQSM's supported by the Supersites benefits both ozone and aerosol implementation programs. An array of meteorological measurements

¹ Diagnostic activities attempt to determine whether the assessment tools truly recreate physical and chemical processes in the atmosphere. For example, an air quality model often is evaluated with data that are available. Historically, ozone model applications relied on available ground level ozone data. The resulting model evaluation provides some limited confidence that the model works as it should; however, so many coupled chemical and physical pathways potentially create circumstances where accurate ozone predictions result from compensating errors in process characterizations (e.g., underestimated mixing and emissions fields). These errors compromise the model's future predictive ability when applied to various emission scenarios. A diagnostic approach to evaluation would provide measurements of key chemical components in near continuous time to test whether the model is capturing important processes, to ensure confidence in reproducing various emission scenarios. In addition, measurements extending vertically clearly are needed given large spatial gradients that often exist in the atmosphere. The problem is magnified with aerosols, where routine measurements typically are aggregated over 24 hours, whereas significant transient changes in atmospheric mixing and emissions occur over much smaller time periods. Moreover, aerosol model evaluations are challenged by additional complexities of multiple phases and size distributions of varied chemistry.

which capture three-dimensional characteristics of wind velocity, temperature, pressure, relative humidity, and other parameters useful for defining structural features for mixing purposes is especially critical for model evaluation.

- **Application of Receptor² and Observation-Based³ Models.** A suite of source apportionment techniques that relate chemically-spectate ambient data to chemical features specific to particular source emissions will be applied to attempt to delineate the principal source categories and their quantitative impacts at a specific location. Examples of these techniques include the Chemical Mass Balance Model version 8 (CMB8) and various multivariate approaches, such as multiple linear regression, constrained factor analyses (e.g., SAFER/UNMIX), and Principal Component Analysis. The multivariate approaches require the simultaneous use of many ambient samples, but have the potential of avoiding the need for externally-supplied source profiles, as in the case of CMB8. An additional promising but relatively unexplored source apportionment technique is the use of the chemical and morphological features of the individual particles composing an ambient PM sample, as determined from scanning electron microscopy (SEM) analysis. All of these techniques are limited by the richness of available data. Thus, while routine 24-hour averaged samples of chemical constituents can be used to drive these techniques, it is often preferable to use data aggregated over smaller time periods that reflect diurnal emissions release patterns (e.g., mobile sources) and/or meteorologically stable periods. Also, specific chemical markers that associate closely with known sources enhance source apportionment analyses. The Supersites are expected to provide specific organic compound data not routinely collected that should enable apportionment, for example, between sources such as (1) diesel- vs. gasoline-powered vehicles, (2) nominally similar vehicles but with very different emissions characteristics (hot-stabilized vehicle emissions vs. “smokers”), and (3) food processing/cooking vs. biomass-derived emissions.
- **Air Quality Trends and Tracking Progress of Control Programs. A**

² Source apportionment is a very broad term that can extend to the more deterministic Eulerian models which develop source-receptor relationships. Depending on perspectives, there may or may not be much delineation between the terms source apportionment and source-receptor. For clarification, we will use source apportionment to refer to the more observation driven approaches such as chemical mass balance receptor models.

³ Observational Models require ambient data as explicit inputs to drive model calculations; more deterministic air quality simulation models are driven principally through emissions and meteorological input fields. A class of observational methods infer “preferred” general control strategy approaches based on atmospheric chemistry conditions. For example, the smog production algorithms (Blanchard et al., 1992) and the Georgia Tech OBM (Cardelino and Chameides, 1995) indicate whether ozone is preferentially reduced through reductions in Nitrogen oxides or volatile organic compounds, rather than identifying a particular source category such as a coatings operation or specific combustion source. Techniques for identifying the relative importance of ammonia and nitrogen dioxide for nitrate aerosols recently have been developed (Blanchard..., 1997).

continuous tracking of air quality is required as a basic accountability measure of ensuring that 1) planned emission strategies are implemented as designed, and 2) such programs achieve acceptable results. Such accountability is absolutely required given so many predictive uncertainties in atmospheric processes and related technical tools as well as unknowns associated with forecasting economic and demographic change. The atmosphere includes complex nonlinearities⁴ as well as negative and positive feedbacks⁵ that demand constant assessment of the effectiveness of emissions strategies. [These same concerns demand that AQSM's undergo diagnostic level testing.] The routine chemical speciation program will provide the basic information to test programmatic effectiveness. While the routine program should provide a measured signal indicating program effectiveness, it is unlikely that routine programs will be capable to explain the "why" or "how" behind unfulfilled program objectives, given the complexity of aerosol and oxidant systems. Clearly, the long term availability of detailed chemical, size and time-resolved data enhances our ability to diagnose the successes and failures of implemented programs, and ensure needed adjustments to optimize air quality management. Similarly, the uncertainties associated with relating specific PM mass components to adverse health impacts demand that a long term tracking system exist to retrospectively review associations as research provides more insight into the relationship between specific PM components and health effects.

⁴ A proportionate reduction in a pollutant precursor does not result in a proportionate reduction in the targeted pollutant

⁵ A reduction in a precursor can lead to an increase or decrease in targeted pollutant level

Appendix C: Overview of National PM_{2.5} Monitoring Networks

The current planned scope of the national PM_{2.5} network consists of three major components: mass monitoring, routine chemical speciation, and special study areas termed “Supersites.” In very broad terms, the network, as a whole, supports three principal regulatory objectives: 1) determining nationwide compliance with the NAAQS; 2) State Implementation Plan (SIP) development (e.g., source attribution analysis and air quality model evaluation; and 3) tracking trends and progress of emissions reduction strategies. The EPA recognizes that, with care in design and execution, components of this program can also provide significant support for priority research needs. The following brief description is intended to provide background for understanding the context and relationship between these components and between them and EPA’s research program. The attached table outlines for each category below a synopsis of the budgeted number, major purposes, and potential flexibility for integration with PM research programs.

MASS MONITORING (1100)

1. **Core mass monitoring (850).** Approximately 850 NAMS/SLAMS sites, required according to EPA guidance to the States, will be dedicated to mass monitoring. A breakdown of these 850⁶ sites includes 750 required for NAAQS compliance and 100 sites for characterizing background and transport. The regulation requires a continuous sampler to be collocated with an FRM/FEM at the 52 largest cities (greater than 1,000,000 population).
2. **Mass samplers for spatial averaging and special purpose monitoring (SPM)(200).** Roughly 200 additional sites to accommodate spatial averaging⁷ and special purpose monitoring needs are expected to be deployed. The SPM sites are those established to identify unique source location or communities, and are not required to be compared to the NAAQS if operating less than 2 years (or a sampler without FRM/FEM designation).
3. **Continuous monitoring (50).** In addition to the required collocated 52 continuous monitors, plans include deployment of an additional 50 continuous samplers. Collectively,

⁶ EPA network guidance (40CFR58) requires 850 NAMS/SLAMS sites; however, 100 of those sites are to be designated as background or transport sites (2 per State) which can use the IMPROVE sampler, which is not designated as an FRM/FEM and therefore would not be used for NAAQS comparisons.

⁷The annual PM_{2.5} standard is specified as reflecting an area-wide distribution or spatial average of a representative single monitor or the average of multiple monitors. States have requested additional monitors to provide for spatial averaging.

at least 100 continuous samplers will be deployed, and probably more, since the States can elect to purchase and operate continuous samplers for sites designated as special purpose monitoring.

Principal objectives for mass monitoring:

- (a) FRM/FEM samplers and NAMS/SLAMS. The primary objective for mass monitoring, especially the designated NAMS/SLAMS¹ sites are for comparison to the PM_{2.5} NAAQS. In addition, 100 NAMS/SLAMS will serve as background and transport sites, integrated with other efforts such as IMPROVE, to characterize regional transport and background concentrations.
- (b) Continuous samplers. Continuously operating samplers will provide a real time estimate of PM_{2.5} levels and allow for input into public information displays (similar to current ozone mapping efforts that reach local weather forecast venues) as well as the Pollutant Standards Index (PSI). Other objectives for continuous samplers include developing statistical relationships with FRM/FEM's to serve as potential surrogates for compliance indicators, and characterizing diurnal patterns of exposure and emissions.
- © Special Purpose Monitors (SPM's). The SPM's samplers are intended to provide flexibility for State and local agencies to investigate areas that may have exceedances without the repercussion of regulatory requirements associated with a NAAQS violations. The purpose of SPM's is to encourage monitoring where it might otherwise be discouraged due to fear of associated regulatory requirements. The SPM's are expected to be located in unique or rural communities subject to localized sources, or enhance the regional/background/transport network to better characterize multiple spatial scale interactions. Samplers for SPM's purposes can be FRM/FEM that operate less than 2 years, or non-FRM/FEM samplers. Many State and local agencies are expected to operate continuous samplers within the classification of SPM's sites.

ROUTINE CHEMICAL SPECIATION (300).

The routine chemical speciation program consists of two components: 50 required NAMS, and up to 250 additional sites (EPA's contribution to the IMPROVE program technically is similar to the routine speciation program but addressed separately due to budget considerations). The major purpose of these sites is to assess long-term trends in major PM_{2.5} components, as well as to provide useful information for source apportionment, evaluating current and future control programs, and health risk assessments.

1. **NAMS (50)**. The regulation requires 50 speciation sites across the country, located mostly in urban areas (e.g., all PAMS cities will have a speciation site). These 50 sites will be designated as NAMS and will follow sampling and analysis protocols similar to the

existing Interagency Monitoring of Protected Visual Environments (IMPROVE) program. Filter sampling techniques (Teflon, nylon and quartz media) for 24-hour periods will be analyzed for principal mass components: most elements through X Ray Fluorescence; major ions through Ion Chromatography/Colorimetry (nitrates, sulfates, chloride/ammonium); and organic and elemental fractions of carbon through Thermo analysis. The sampling methodology and frequency (1-in-6 day or greater) are being evaluated in light of peer review comments. Prescriptive protocols for sampler selection, analytes, and sampling frequency will be adhered to ensure national consistency across space and time.

2. **Other “Routine” Speciation Sites (250).** In addition to the NAMS, resources are expected to be available to support up to 250 additional sites. These sites will be less prescriptive than the NAMS and will be subject to a balance among competing needs for national consistency (50 sites are not adequate to characterize the U.S., suggestions for more frequent sampling), and flexibility to address local-specific issues such as winter time wood smoke, or the need to support related scientific studies, which might require more intensive seasonal sampling and analysis. This component of the program does provide true flexibility for State and local agencies. Certain States (e.g., California) have expressed an interest in establishing more advanced methods capable of in-situ, near continuous measurements of principal species. Given the flexibility of this component of the national program, substantial opportunity exists to interact with the health and atmospheric chemistry research communities. With the exception of the Supersites program, however, all of these components are funded by State Grants, which provide hardware and related capitol costs, laboratory analyses, and salaries for State and local agencies to operate the network. Consequently, the dialogue must involve EPA, State and local agencies, and the research community.
3. **IMPROVE Sites (108).** In addition to 30 existing EPA supported sites, 78 new IMPROVE sites are being added, in or near Class I Federal areas (e.g. national parks and wilderness), to address the requirements of the forthcoming Regional Haze regulations. These sites conduct speciation sampling similar to the 50 NAMS, but on a 1/3 day sampling interval. These sites are considered as part of the entire PM_{2.5} National network, recognizing that the technical connections (e.g., sources/ambient characterizations, measurement techniques) between PM_{2.5} and visibility require integration. Although funded through State Grant funds, this program is managed by the IMPROVE Steering Committee, and most of the technical work conducted by Universities and the Federal Land Managers.

SUPERSITES (4-7). See main text.

Scientific Review of Network Components

The use of PM_{2.5} mass as an “indicator” for PM standards was recommended by the Clean Air Scientific Advisory Committee (CASAC) at the conclusion of their review of the scientific criteria and standards. Both the Federal Reference Method for measuring PM_{2.5} mass and EPA’s guidance for establishing the mass compliance network were peer reviewed by the Fine Particle Technical Monitoring Subcommittee of CASAC in 1996. The more recent plans for speciation measurements, continuous monitors, and Supersites are in partial response to the subcommittee’s recommendations for monitoring beyond 24-hour PM_{2.5} mass. The approach for the required speciation monitoring network was recently reviewed by an expert scientific panel in Seattle. The approach and objectives for the super site program will be the focus of a major 2-day workshop in July. A workshop planning group, including a number of recognized scientific experts in health, exposure, atmospheric sciences, and monitoring met in May and is continuing to develop materials for the program. In addition to providing periodic updates on this program to the NAS panel, EPA intends to present its approach for integrating the “routine” speciation network with the super site monitoring and research programs for review by the Fine Particle Monitoring Technical Subcommittee of CASAC in the Fall.

List of Acronyms

PM_{2.5} = Particles with an aerodynamic diameter less than or equal to a nominal 2.5 micrometers

PM = Particulate Matter

EPA = Environmental Protection Agency

NAMS = National Air Monitoring Station

SLAMS = State/Local Air Monitoring Station

NAAQS = National Ambient Air Quality Standards

FRM = Federal Reference Methods

FEM = Federal Equivalency Methods

SPM = Special Purpose Monitoring

IMPROVE = Interagency Monitoring of Protected Visual Environments

PSI = Pollutant Standards Index

Overview of National PM_{2.5} Network

Site Category	Projected Number	Major Purpose	Potential Flexibility for Research
Core Sites	850 FRM/FEM measure PM _{2.5} mass. Also 50 collocated monitors measure continuous mass	Minimum required for designations. FRM and network design peer reviewed by CASAC. Continuous required for PSI reporting.	Limited. States follow EPA guidance on location according to population, other factors. Frequency of sampling could be adjusted at some.
Spatial Averaging/ Special Purpose	200 FRM/FEM, other	States requested additional monitors for spatial averaging for attainment designations. SPMs limited duration (<2 yr), e.g. source attribution study	Locations determined by States according to local circumstances. SPM might be adjusted to accommodate research
IMPROVE	100 additional IMPROVE monitors	Supports regional haze rules in class I areas and PM _{2.5} transport assessment. Chemical speciation.	Limited to class I areas.
Chemical Speciation	300 sites with "routine" chemical analyses	Trends, source attribution of major chemical species, for source apportionment, risk assessment. Regional variations encouraged.	Substantial flexibility to accommodate health and other research subject to resource limitations on frequency.
Continuous	50 additional continuous PM _{2.5} mass monitors	PSI reporting and further delineation of source/exposure patterns	Substantial flexibility to support exposure studies.
Total	1500 Sites		

In addition 4 to 7 Supersites not included in above with research grade instrumentation will be established for health risk and source assessment work integrated with research program. The design of this program is fully flexible for incorporation into other priority scientific research on PM.

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PM WORKSHOP STEERING COMMITTEE

Co-Chairs

Dan Albritton	NOAA Aeronomy Laboratory, Boulder, CO
Dan Greenbaum	Health Effects Institute, Cambridge, MA

Extramural Members

Kurt Anlauf	Environment Canada, Downsview, Ontario, Canada
Steve Cadle	General Motors Research and Development Center, Warren, MI
Glen Cass	California Institute of Technology, Pasadena, CA
Jeffrey Cook	California Air Resources Board, Sacramento, CA
Ken Demerjian	State University of New York at Albany, Albany, NY
Howard Feldman	American Petroleum Institute, Washington, D.C.
Susanne Hering	Aerosol Dynamics Inc., Berkley, CA
Petros Koutrakis	Harvard School of Public Health, Boston, MA
Paul Lioy	Rutgers University, Piscataway, N.J.
Joe Mauderly	Lovelace Respiratory Research Institute, Albuquerque, NM
Jim Meagher	NOAA Aeronomy Laboratory, Boulder, CO
Lucas Neas	Harvard School of Public Health (now at EPA, RTP, NC)
Jonathan Samet	Johns Hopkins University, Baltimore, MD
Pradeep Saxena	Electric Power Research Institute, Palo Alto, CA
Richard Schlesinger	New York University, Tuxedo, NY

EPA Affiliates

John Bachmann	Dan Costa
Rich Scheffe	Linda Sheldon
Paul Solomon	John Vandenberg
Jim Vickery	Russell Wiener
William Wilson	

PROPOSED WORKSHOP AGENDA

July 22

- | | | |
|--------------|---|--|
| 8:30 | Welcome and Overview | Dan Albritton &
Dan Greenbaum |
| 9:00 | EPA Vision for PM Measurement Program | Gary Foley |
| 9:20 | Overview of NAS report -
<i>Research Priorities for Airborne Particulate Matter</i> | Dan Greenbaum |
| 9:45 | The EPA PM Chemical Speciation Network | Petros Koutrakis |
| 10:15 | PM Measurement Workshop Report | Peter McMurry
C. S. Kiang |
| 10:45 | Break | |
| 11:00 | Panel Discussion¹ - Moderated by Dan Albritton (Research Monitoring objectives - charge to breakout groups) | |
| | Health Effects | Joe Mauderly |
| | Exposure Assessment | Paul Lioy |
| | Source / Receptor Relationships | Glen Cass |
| | Accountability | Ken Demerjian |
| | PM Measurement Methods | Susanne Hering |
| 12:30 | Lunch Break | |
| 2:00 | Breakout groups convene separately | |
| 5:00 | Breakout groups adjourn | |

July 23

- | | | |
|--------------|--|--|
| 8:00 | Breakout groups reconvene to finalize² reports | |
| 9:30 | Breakout Discussion Leaders summarize major changes to the draft workshop report / discussion | |
| | Health Effects | Joe Mauderly |
| | Exposure Assessment | Paul Lioy |
| | Source / Receptor Relationships | Glen Cass |
| | Accountability | Ken Demerjian |
| | PM Measurement Methods | Susanne Hering |
| 12:30 | Lunch Break | |
| 2:00 | Summary of findings, commonalities, priorities and future actions | Dan Albritton &
Dan Greenbaum |
| 3:30 | Adjourn | |

Notes

1. Each Discussion Leader will briefly outline the major points in the material provided in their section of the draft plan. A list of key issues / questions to be discussed in the breakout session will also be presented.
2. The Discussion Leader and the Rapporteurs for each area will meet after the breakout sessions on day 1 to develop a summary of the deliberations. The members of each breakout session will meet early on Day 2 to refine this summary before it is presented to the entire workshop.

Discussion Leaders and Rapporteurs For Breakout Sessions

Health Effects

Discussion leader - Joe Mauderly
Rapporteur - Richard Schlesinger
Rapporteur - Lucas Neas

Exposure Assessment

Discussion Leader - Paul Lioy
Rapporteur - Petros Koutrakis
Rapporteur - Linda Sheldon

Source / Receptor Relationships

Discussion Leader - Glen Cass
Rapporteur - Pradeep Saxena
Rapporteur - Howard Feldman

Accountability

Discussion Leader - Ken Demerjian
Rapporteur - Jeff Cook
Rapporteur - TBA

Measurement Methods

Discussion Leader - Susanne Hering
Rapporteur - Kurt Anlauf
Rapporteur - Steve Cadle

INTRODUCTION

On July 18, 1997 the U.S. Environmental Protection Agency (EPA) revised the National Ambient Air Quality Standard (NAAQS) for particulate matter (PM) [EPA, 1997a], changing the metric from PM_{10} (particles with aerodynamic diameters less than 10 micrometers) to $PM_{2.5}$ (particles with aerodynamic diameters less than 2.5 micrometers). In taking this action the EPA cited epidemiological evidence linking significant human health impacts (mortality, hospital admissions, respiratory illness) to ambient particulate levels below the previous standard. In addition to this regulatory initiative, which was focused on improving public health, the EPA has proposed new Regional Haze Regulations [EPA, 1997b] to protect and improve visibility in the 156 mandated Class I areas (National Parks and Wilderness Areas) of the country. Fine particles are the single greatest contributor to visibility impairment in these pristine areas of the country.

In Canada PM is regulated through an Ambient Air Quality Objective for Total Suspended Particulates (TSP). The framework for future additional/replacement regulation on PM is presently being reviewed with an emphasis on developing a regulation with a strong science foundation. Options for both PM_{10} and $PM_{2.5}$ standards are under discussion. Mexico currently has ambient air quality standards for both TSP and PM_{10} .

A i r b o r n e

particles may

have many

sources and

c o n t a i n

hundreds of

inorganic and

thousands of

o r g a n i c

components.

Sources, size,

c h e m i c a l

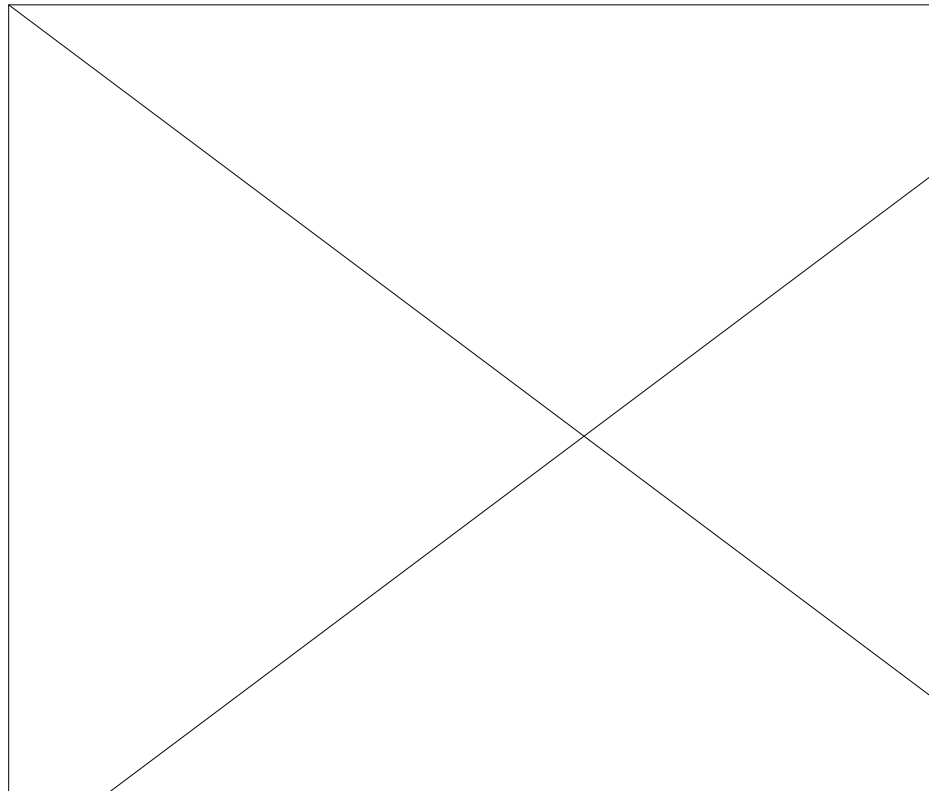
composition

a n d

atmospheric

b e h a v i o r

divide particles



into Òfine and coarse modesÓ with a split at about 2.5 μm . Much of the fine particle mass found in the atmosphere is not emitted by any one source but rather is formed in the atmosphere as a result of chemical conversion of man-made and natural emissions.

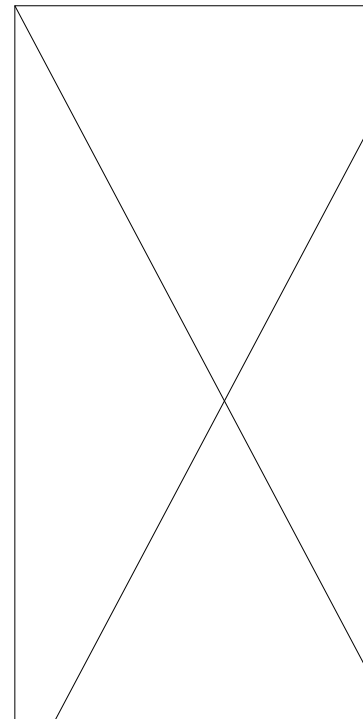
By mass, fine particles are primarily sulfate, nitrate, ammonium ions, carbon soot, and organics, as well as mineral dust in some locations. Course particles in the range 2.5-10 μm are largely deposited in the nasal-pharyngeal areas; particles smaller than 2.5 μm may reach the lungs.

Ambient fine particulate levels vary greatly both regionally and seasonally. Sulfate constitutes a significant fraction of the fine particle mass in the East, with nitrate and carbon (elemental and organic) playing lesser roles. However, in the West, the fine particle mass is dominated by nitrate and carbon with sulfate making a smaller contribution (Figure 2), reflecting differences in emissions in the two regions. Regional background fine particle levels (and associated visibility impairment) are typically highest in the eastern U.S. in the summer months, while levels typically peak in the winter months in southern California. Average visual range in most of the western U.S. is 60-90 miles, or about one-half to two-thirds of what it would be without pollution. In most of the East, the

average visual range is less than 18 miles, or about one-fifth of the visual range that would exist under natural conditions.

Indications from epidemiological studies of an association between ambient particles and human health endpoints suggest decreased emissions could lead to reductions in premature

mortality and morbidity from cardiovascular and respiratory causes. However, the biological mechanism(s) by which particulate matter at low ambient levels could cause mortality and morbidity is uncertain.



The EPA has delayed implementation of the new PM NAAQS until after the next periodic review of the Federal air quality standards, which is scheduled for 2002. This delay could allow time for the national PM monitoring network to be put in place and acquire a measurement record that is long enough (3 years) to perform attainment/nonattainment designations. This period also provides an opportunity to clarify the associations between PM and health effects and to improve our understanding of the processes that influence PM formation and distribution in the atmosphere, both of which are essential to effective mitigation.

The U.S. Congress, in response to public health concerns and recognizing the uncertainty associated with some key aspects of the science, has provided support for a major PM research initiative. The National Research Council (NRC), at the request of the EPA Administrator, has established an independent committee of experts to identify the major priorities for such an initiative. The Committee on Research Priorities for Airborne Particulate Matter has produced the first in a series of

four reports on PM research priorities, *Research Priorities for Airborne Particulate Matter I: Immediate Priorities and a Long-Range Portfolio* [NRC, 1998]. In this report the committee identifies the areas of PM research that deserve the greatest emphasis and provides a conceptual design for a policy-relevant research program.

The EPA/NARSTO PM Workshop

As part of this research initiative EPA, The North American Research Strategy for Tropospheric Ozone and Aerosols (NARSTO), and other interested stakeholders are planning a program of detailed measurements of atmospheric particles and important co-pollutants. Measurements will be made in areas representative of those where particles are expected to create potentially significant health risks and therefore, pose representative control-strategy issues facing those charged with implementing the PM_{2.5} NAAQS. These measurements will be made as part of a larger, multi-purpose PM monitoring system being implemented by EPA and state and local environmental agencies (see Appendix A). Although this larger system is designed primarily to apply the Federal Reference Method (FRM) for PM_{2.5} to determine compliance with the NAAQS, there is substantial flexibility in portions of this system to complement and augment the more detailed research measurements at more limited sites that are discussed in this document.

The PM Workshop Steering Committee, comprised of experts from the atmospheric, exposure and health effects scientific communities, was convened to provide some initial ideas on the basic elements of a PM research measurement program. The Committee was also tasked with the design of a workshop where these ideas could be reviewed and expanded by the views of the broader PM research community and stakeholders. The Workshop Steering Committee is co-chaired by: Dr. Daniel Albritton, National Oceanic and Atmospheric Administration, Boulder, CO and Mr. Daniel Greenbaum, Health Effects Institute, Cambridge, MA. A complete listing of the Steering Committee members is provided above. The Committee met in Research Triangle Park, North Carolina on May 19, 1998 and developed the design for this workshop and the material provided in this report. This concept paper is intended as a starting point for discussion that will allow the Workshop attendees to focus on the difficult issues that are most critical to the design of a successful research program.

This Workshop is being held to discuss the views of the atmospheric, exposure, and health-effects scientific communities regarding measurements at chemical speciation sites and special-study "super sites." The discussions will focus on scientific questions and issues, preferably in the form of

testable hypotheses. The location, duration, and kinds of measurements that are needed to accomplish such research will also be discussed. **The goal of this Workshop is to identify the key components and design parameters for a comprehensive measurement program to characterize ambient particulate matter and important co-pollutants in a way that optimizes information for multiple disciplines, including source apportionment; modeling, health and exposure study; and risk assessment.**

The PM Workshop Steering Committee feels strongly that the term "super site", when applied in the context of this workshop, should be thought of in the broadest possible terms. A comprehensive array of state-of-the-art measurements performed at a limited number of sites, as well as associated theory and modeling, could represent one possible component of the program. A larger network employing a more limited set of focused measurements or intensive field campaigns that utilize instrumented aircraft and ground-based measurements may also play a role. The focus should be on developing the best possible research program using the measurement tools and techniques that will be most effective, and designing the program to be implemented in close cooperation with the EPA and the state and local environmental agencies

In preparation for the Workshop, the members of the Steering Committee were asked to provide some initial thoughts on the design of a PM measurement research program from the perspective of their individual areas of interest. These areas of interest fall into three broad categories:

Health effects

1. Personal exposure assessment
2. Source/receptor relationships

The panel identified two additional cross-cutting issues that relate to all of the areas listed above:

3. Accountability determination of the effectiveness of management strategies. Are the emission management programs producing the air quality improvements we expect? If not, why not?
4. Evaluation and development of PM measurement methods. How good are the methods we are currently using to characterize PM? What improvements are needed?

In order to facilitate the integration of the needs and issues identified by the various represented disciplines, the authors structured their ideas around the following set of questions:

What are the major science questions/hypotheses? From a process and receptor-oriented perspective, what is the state (chemistry, size, and phase over time) of aerosols and the relevant formation, maintenance, and removal processes responsible for the existing (and future) state? What measurements are needed to diagnose and evaluate the sophisticated air quality simulation models that simultaneously predict oxidant and aerosol concentrations? Such formulations should recognize the hypotheses currently under consideration by the health effects community regarding linkages among exposure to aerosol characteristics, biological mechanisms, and the observed health effects due to inhalation of fine particulate matter. These hypotheses will be first-order guidance to the atmospheric and exposure communities regarding the types of measurement data that could be used to differentiate among the various hypotheses as to biological causal mechanisms. Further, these hypotheses will guide the formulation of relevant atmospheric science questions, such as potential linkages between source types and concentrations/exposures.

•What is to be measured? This question encompasses the attributes of the aerosols including mass, chemical composition and physical characteristics (size and shape). Also included are chemical precursors, intermediates, sinks, other pollutants, tracers, and meteorology. The geographical location may also influence the type of measurements that are selected.

•Where are the measurements to be made? How many sites/study areas/airsheds need to be studied? How should these sites/study areas/airsheds relate to the monitoring networks that are in place or are being implemented by the EPA and state and local environmental agencies? What are the implications of these research needs for the implementation of the more flexible portions of the larger monitoring network (e.g. the supplemental speciation and continuous monitoring sites)? Are the objectives better served by measurements made in urban areas where the greatest population exposure occurs or in rural areas where the influence of the natural background can be documented and the regional nature of the problem studied? Will the program objectives only be served by a mixture of urban and rural sites? Should the sites be paired? How do outdoor measurements relate to indoor and personal exposures to particles of ambient and other origin? How is spatial representativeness determined?

•When will the measurements be made? What are the recommended interval, frequency, seasonality and duration of the measurements? What are the relative merits of continuous measurements versus a series of intensives?

Some initial thoughts on areas of overlap, where specific measurements and measurement strategies support multiple objectives, are provided in the last section of this document. The identification of these areas of overlap is a primary focus of the Workshop. The breakout sessions at the Workshop will, while disciplinary by topic, deliberately have some mix of other interests to start this process. And, subsequently, the identification of areas of overlap will be a major component of the second plenary.

The following descriptions of the key questions and information needs for each area and issue are provided as initial thoughts to focus the discussions at the July 22-23, 1998 Workshop. Each of these will be discussed in some detail at breakout groups on the first day, to ensure that they are accurately describing the needs in each area. Then, during the evening, and in discussions on the morning of the second day, the common and disparate needs across these areas will be identified, leading to the development of an integrated suggested program of research-related measurements as a final outcome of the workshop.

MEASUREMENT NEEDS RELATED TO HEALTH EFFECTS

Prepared by Joe Mauderly, Richard Schlesinger and Lucas Neas

There are statistical associations between short-term increases in ambient particulate material (PM) and daily mortality and morbidity. Mortality appears to have as a basis both respiratory and cardiac causes and seems to occur primarily among elderly individuals, presumably those having pre-existing respiratory and cardiac disorders. Morbidity is most significant for respiratory conditions, as indicated by increased clinic access, hospitalization, medication usage, and reductions in lung function. There are also indications, although based on fewer data, of statistical associations between average long-term PM levels and increased mortality rates. Deaths from cardiorespiratory causes and lung cancer appear to be most strongly associated with long-term PM exposure.

There remains considerable uncertainty regarding the link between ambient PM and health effects. Although it has not been proven that causality is attributable to PM alone, current evidence strongly suggests that PM is at least a key factor, if not directly causal. EPA, the National Academy of Sciences, and other groups have emphasized the need for research to better understand the links between PM and health effects. Among the several categories of research identified, two interrelated issues are consistently listed as high priority: a) understanding the physiochemical characteristics of PM most closely related to the observed health effects; and b) understanding the biological mechanisms by which these effects occur. It is thought unlikely that a single unifying hypothesis will be generated to explain the PM-health linkage. Rather, it is generally accepted as most likely that multiple toxic species act by several mechanistic pathways to cause the range of health effects that are observed.

The designing of atmospheric measurement studies that will serve to assist epidemiological and toxicological studies in evaluating health effects from exposure to ambient PM requires some knowledge of the characteristics of ambient PM that are likely responsible for the observed statistical associations noted above. While the database in this regard is currently not clear, there are numerous candidate hypotheses about the characteristics of ambient PM responsible for health effects and the mechanisms by which these characteristics result, either directly or indirectly, in these effects. The ten general hypotheses listed below do not comprise an exhaustive list, but encompass those invoked most frequently during and after the recent review of the PM NAAQS. There is evidence of varying

degrees to support the importance of each of these hypotheses, but there are also counter-arguments. Regardless, the following unprioritized ten hypotheses provide a starting point for structuring sampling strategies for health research.

Current hypotheses of PM characteristics related to health effects

1. PM Mass Concentration

The epidemiological data indicating that ambient PM causes a range of health effects are based on the correspondence between measures of PM mass concentrations and measures of health responses. Moreover, the similarity of the PM mass-health relationships in many locations encompassing different climates and pollutant composition suggests that mass is a useful unifying PM measure. Because it is well known that: a) PM includes materials having diverse physical-chemical characteristics; and b) not all particulate materials are of equal toxicity, it is generally accepted that some portions of PM are more important in producing adverse health consequences than are others. It cannot be ignored, however, that most of the current concern arose from associations of health endpoints with mass concentration, not chemical composition. There are indications that fine PM (e.g., PM_{2.5}) is more potent than coarse PM on a mass concentration basis, but other than that, our existing information is based on mass as measured by current compliance monitoring procedures.

2. Metals

Metals, particularly reactive transition metals such as vanadium, copper, iron, platinum, etc., are known from previous toxicological studies to have cytotoxic and inflammatory properties. Because these metals are ubiquitous constituents of ambient PM, they have been hypothesized to be important in the observed effects. The metals hypothesis resulted in, and has been supported by, numerous studies showing that residual oil fly ash (ROFA), usually at very high doses, is toxic to cells and the lung, can cause physiological abnormalities in animals when instilled or inhaled, and can cause death in compromised animals. The effects are primarily associated with the soluble metal fraction and may be related to the ability of these metals to catalyze production of free radicals in tissues. Support for this hypothesis is provided by combined epidemiological and toxicological studies showing that the health effects from PM emitted from a steel mill in the Utah Valley were most likely attributable to its metal content. The extent of the contribution of

metals to the effects of other ambient PM is unknown, although there is growing information suggesting that metals are at least among the chemical constituents of concern.

3. Acids

Acid aerosols, and acidic PM, have been known for many years to have toxic properties in laboratory studies, largely corresponding to the concentration and amount of H⁺ delivered to respiratory surfaces. There is some, but less, evidence for the role of airborne acids in health effects from exposure to ambient air pollution. The potential role of acids in the observed PM effects is debatable on the basis of neutralization, etc., but acids and acidic PM constituents clearly still remain as a potential hypothesis.

4. Organic Compounds

Ambient PM contains organic compounds, as particles formed from volatile and semivolatile organic vapors, as organic material condensed in the atmosphere on solid particles, and as the adsorbed organic fraction of soot. It is known from previous work that this diverse class of material contains irritants, mutagens, and carcinogens. It is speculated, but largely unknown, that organic constituents may act as irritants, or allergens (alone or in combination with other constituents). It is known that this class of material in ambient samples produces mutagenicity and can cause cancer in high-dose laboratory studies. Perhaps the strongest support for the importance of organics comes from the finding that lung cancer mortality bore as strong a statistical relationship to increased mortality rates as cardiorespiratory mortality in the Six-Cities Study [Dockery *et al.*, 1993].

5. Ultrafine PM

For this purpose, "ultrafine PM" is defined as particles having diameters of 100 nanometers (0.1 µm) or less. Ultrafine ambient PM is generated largely from combustion sources and is universally present. The majority of ambient particle number is contributed by ultrafines. On the other hand, ultrafine particles are generally short-lived due to agglomeration into larger particles and comprise only a small portion of total ambient PM mass. Laboratory studies have shown that, for deposition of a given material in the lung, toxicity tends to increase as particle size decreases. This is plausible on three bases: a) the finer particles penetrate more readily into cells and through tissue barriers; b) the finer particles have greater surface area per unit of mass, and many toxic reactions presumably occur at the surface; and c) finer particles dissolve more rapidly than do larger particles, thus enhancing the bioavailability of solubilized agents. None

of the epidemiological studies leading to the present PM concerns included measures of ultrafine PM, and the importance of ultrafines remains largely speculative. However, increasing attention is being given to ultrafines, and there is a growing recent epidemiological database suggesting that this fraction may be important.

6. Biologicals

It is often overlooked that a portion of ambient PM is of biological origin, including bacteria and viruses, bacterial endotoxins, pollens, plant and animal detritus, and proteins of diverse origin adsorbed to PM of non-biological origin. Some of these materials, such as pollen, are larger than PM_{10} and are not included in reported PM levels. This class of materials is of concern for three reasons: a) infectivity; b) cytotoxicity and inflammatory potential; and c) allergenicity. Ambient airborne biological PM or PM constituents have received little attention during the recent PM discussions, but should not be ignored. Their involvement in the observed effects is plausible.

7. Sulfate and Nitrate Salts

It is known from toxicological experience that sulfate and nitrate salts can be toxic. The majority of laboratory research has focused on the acidic species, and the health concerns overlap those described for acids above. A wide range of inorganic and organic sulfur and nitrogen compounds has irritating, cytotoxic, and mutagenic properties. Sulfates and nitrates may prove to be important PM constituents.

8. Peroxides

Cellular injury from reactive oxygen species has been demonstrated in toxicological studies of particles and is a plausible mechanism for at least some ambient PM effects. Peroxides comprise one of several chemical species causing oxidant injury and may be important in PM effects. It will be useful to characterize the peroxides associated with ambient PM, as well as the generation by PM of oxidant species in biological media.

9. Elemental Carbon (soot)

Elemental carbon is often used as a marker for soot in ambient PM. Elemental carbon particles, such as carbon black, have been shown, in laboratory studies, to cause tissue irritation and the release of toxic chemical intermediates from scavenger cells. However, the principal concern is for the health effects of soot, which is comprised of an elemental carbon matrix with adsorbed organic compounds, metals, and acids. Soot has irritant, mutagenic, and carcinogenic properties

that vary with delivered dose. It is assumed likely that soot is an important class of ambient PM regarding health effects, and it is plausible that it could exert both short-term (irritant) and long-term (carcinogenic) effects. It is useful, therefore, to measure elemental carbon as a marker for soot.

10. Co-Pollutants

Although not a hypothesis regarding PM composition, the issue of co-pollutants, especially gases, continues to be considered very important. First, because air pollution is always a mixture, it remains possible that the "critical" pollutant species vary with PM in concentration, but may not, or not always, be PM itself. Second, it is plausible, and perhaps likely, that the observed effects result from exposure to a combination of PM and other pollutants. It cannot be ignored that the present PM-health associations may result from the fact that PM is the most robust indicator of "dirty air". While our present knowledge points toward an important role for PM itself, it is very important to examine the potential role of co-pollutants.

Useful air parameters

The description above of current hypotheses related to PM health effects provides an indication of some of the parameters that would be most useful to measure in the ambient PM sampling sites. While total mass concentration of $PM_{2.5}$ and PM_{10} would be assessed for compliance purposes of fine and coarse particle modes, it would be important to also measure particle number for evaluation of ultrafine particles at the various sites. Particulate-associated acidity should be measured, as should metal content, especially for the transition metals and biological aerosols occurring within the $PM_{2.5}$ and PM_{10} modes. Regarding co-pollutants, perhaps the gaseous co-pollutants of most concern based upon current hypotheses would be ozone and sulfur dioxide.

Criteria for selecting airsheds

If the sampling strategy is based upon the current hypotheses above, then the criteria for selection of airsheds must involve evaluation of regions within which ambient PM has the necessary

characteristics to evaluate these hypotheses. However, since effects of PM may be due to multiple components including gaseous co-pollutants, this selection is quite difficult.

As a first cut related to sites, there should clearly be regional sites, since air chemistry can differ in different parts of the country. For example, southern California aerosol has a significant particulate nitrate component with little sulfate, while the eastern cities have more sulfate. Thus, it would seem appropriate to have sampling performed in southern California and a northeastern city. Other sites could be in areas where there is little contribution of acidic particles, such as the Utah Valley, and areas where a significant portion of ambient PM is natural in origin. The issue of co-pollutants is difficult to deal with in site selection. However, areas with significant ozone should be selected for comparison to areas with lower levels, but the characteristics of PM in these sites would also likely differ. This could make it hard to determine the contribution from gaseous co-pollutants, a problem common to many epidemiological studies. In order for a co-pollutant to confound an association between PM and health outcomes, that co-pollutant must co-vary with the exposure of interest. If the co-pollutants are from the same source, the issue is not one of confounding associations; rather, it is the ability of health effects studies to distinguish between highly correlated covariates. The key is to identify monitoring site areas or sampling time periods where the correlation between such co-pollutants is relatively low.

Aside from the designs that use repeated measures of continuous variables, epidemiologic studies require large numbers of exposed individuals. The use of a single central monitoring site to characterize the exposures of an entire urban area is appropriate whenever short-range spatial gradients are low and indoor-outdoor ratios are high. Misclassification of exposure that is nondifferential with respect to outcome will tend to reduce the power of an epidemiologic study by introducing a bias towards the null. Small scale substudies using personal monitoring would be a useful component of any epidemiologic study design, but such studies could not provide the necessary number of subjects and sufficient statistical power to produce reliable associations.

Frequency and duration of measurements

Most likely, the standard six-day measurements from the regulatory networks would not be sufficient. Most likely daily and 12-hr means would provide information that would be more useful in relating atmospheric measures to the proposed hypotheses. The relevant time window for exposure depends upon the induction period and duration of the health outcome of interest. This may be 24-48 hours for mortality or only several hours for incident coronary events. Thus, the monitoring protocol should take into account current hypotheses of mechanisms underlying mortality/morbidity related to ambient PM. These include both pulmonary and systemic, e.g., cardiac, outcomes.

MEASUREMENT NEEDS FOR PERSONAL EXPOSURE ASSESSMENT

Prepared by Petros Koutrakis and Paul Lioy

Most exposure assessment and epidemiological studies have relied on ambient air quality data obtained at a single stationary air monitoring (SAM) site. It is unclear, however, whether ambient concentrations are appropriate surrogates of personal exposures to particulate (PM₁₀ or PM_{2.5}) and gaseous (O₃, SO₂, CO, or NO₂) air pollutants, since people spend the majority of their time indoors. The validity of outdoor particulate concentrations as an exposure measure has been examined in some studies, with most of these studies focusing on the relationship between personal exposures and ambient PM₁₀ and PM_{2.5} concentrations. In these studies, daytime personal PM₁₀ and PM_{2.5} exposures were consistently higher than corresponding outdoor levels. The Particle Total Exposure Assessment Methodology (PTEAM) [Thomas *et al.*, 1993] study, for example, showed daytime personal PM₁₀ exposures to be, on average, 50% higher than corresponding ambient levels, while the Harvard Six City study [Dockery *et al.*, 1993] found mean personal PM₁₀ exposures to be more than 100% greater than mean ambient levels.

Results from recent exposure assessment studies suggest that correlations between personal exposures and outdoor concentrations are only significant for approximately half of the individuals. In addition, significant intra- and inter-personal variability in exposures was found. These differences can be attributed to the differences in activity patterns and home characteristics. As expected, the personal exposures of individuals who spend most of their time outdoors are more closely associated with ambient concentrations. Activity patterns have an additional influence on particulate exposures, as several commonly-performed activities such as cooking, cleaning, and even walking can generate particles. In the PTEAM, Total Human Environmental Exposure Study (THEES) [Liroy *et al.*, 1990] and Six City studies, personal PM₁₀ exposures were found to be significantly higher when individuals performed particle-generating activities such as cooking and cleaning. Housing characteristics also have been shown to affect personal particulate exposures. Weaker winter cross-sectional correlations (as compared to those in the summer) were found in an exposure study conducted in Boston, MA. These weaker correlations can be attributed to seasonal differences in housing characteristics, where wintertime air exchange rates were significantly lower than those in the summer. (Lower air exchange rates provided more time for particles to deposit indoors and for particles to accumulate

from indoor sources.) As a result, associations between indoor and outdoor particle levels, and thus between personal and outdoor levels as well, were weaker in winter as compared to summer.

Less is known about the ability of gases, such as O₃, SO₂, NO₂, and CO, to confound the observed associations between ambient particle concentrations and adverse health effects. To date, no studies have been conducted that address this issue directly; however, even for these pollutants, outdoor concentrations measured at a single SAM site are often poor surrogates for personal gaseous exposures. For instance, in an ozone exposure study conducted in State College, PA, indoor concentrations were found to be the most important predictor of personal ozone exposures, with outdoor concentrations explaining little of the variability in personal exposures. Again, air conditioner use and activity patterns were found to be important determinants of the personal-outdoor relationship. However, other research has shown that, for houses that lack air conditioning, outdoor levels are similar to indoor levels. In general, indoor sources and sinks for O₃ are different from those of particulate matter, which may weaken the associations between personal particulate and gaseous exposures.

In summary, personal and indoor measurements, along with information on activity patterns and microenvironmental characteristics, are necessary to determine human exposures. A number of exposure assessment studies are starting or underway that will investigate factors affecting the relationship between personal exposures and outdoor concentrations. These studies will provide the necessary data to develop exposure models. These models will use information on outdoor concentrations, activity patterns, and home characteristics to determine personal exposures for the measured cohorts. Subsequently, these models will be used to develop population exposure models.

The outdoor component of an exposure study represents a small fraction (less than 10%) of the study costs. However, exposure assessment studies will benefit from the super sites for the following reasons:

1. The super sites will provide information on the diurnal profile of the fine mass and complex chemical components. This information is very important for the development of exposure models.
2. The super sites will use state-of-the-art monitoring techniques, including continuous monitors that will measure a number of physicochemical properties of outdoor

particles. This information will be very useful in determining the contribution of outdoor sources to the indoor environments.

3. Data on the toxic components of fine particles, provided from the super sites, will be used for epidemiological studies and in the design of toxicological studies to identify the health effects and mechanisms of action respectively. As a result of these investigations, future exposure assessment studies will be able to focus on specific chemical, physical, or biological properties of particulate matter.

Air parameters

Initially, an effort that employs super sites should characterize as many particle properties as possible, such as: fine and coarse particle mass, particle number and size distribution, particle morphology, ions (sulfate, nitrate, ammonium, hydrogen, potassium and sodium), elements, organic and elemental carbon, organic carbon species, and biologicals. Also, when possible, measurements of meteorological parameters, such as temperature, dew point, vertical temperature profiles, wind speed, direction and trajectories, etc., should be included in specific super site locations.

The EPA's one-atmosphere approach is sound. The emissions and atmospheric transformation processes that produce ozone and acid rain also lead to the formation of fine particles. Outdoor exposure and, to a lesser extent, indoor exposure are influenced by the pollutant mix that results from the interactions of many pollutants from a variety of sources. Thus, the investigation of relationships between particulate and gaseous concentrations is very critical to our understanding of the sources, transport, fate and health effects of these pollutants. As such, an attempt should be made to measure important gaseous pollutants (such as ozone, carbon monoxide, nitrogen dioxide, sulfur dioxide, and volatile organic compounds). If possible, super sites should be co-located with PM speciation sites, Photochemical Assessment Monitoring Stations (PAMS) sites or other monitoring sites that measure gaseous air pollutants.

Criteria for selecting the sampling location

The analysis of the data from the super sites should focus on the following important issues:

- Characterization of the components of fine and coarse particulate matter and identification of the sources of fine and coarse mass and of specific compounds that contribute to the mass;
- Investigation of the temporal profiles of particle mass and composition; and
- Investigation of the relationship between ambient concentrations and emissions from sources impacting the specific receptor.

The data from the super sites should primarily be used to investigate types of sources and their global emissions, rather than focusing on microscale variability of exposure. A variety of urban environments should be selected for the super site network. The exact number of cities and the specific design will depend to a large extent on the available funding and human resources. However, because of the lack of information on the chemical characteristics in a variety of locales, the initial super measurements should be made using mobile platforms that can be distributed around the country. In order to select these cities, U.S. urban environments can be grouped based on their geographical location, climatic conditions, and types of sources by which they are impacted. Depending on the number of groups and the results using the mobile platform over a period of 3 years, one or two cities can be selected to establish quasi-permanent (~3yr.) super sites per major groups of urban environments.

Fine particulate matter consists mostly of secondary particles, a large fraction of which are formed outside the boundaries of the city or even the state (especially true for Central/Eastern U.S.). Of course there are exceptions, where emissions from local sources such as wood burning or vehicles under certain meteorological and topographical conditions can have an important impact on the local air quality. Particle studies conducted in several eastern U.S. cities (Philadelphia, New York, Washington, Boston) suggest that there may be great similarities in particle composition and that concentrations are highly correlated among these urban environments. In addition, these studies suggest that particle concentrations are relatively uniform and are correlated throughout large metropolitan areas and on subregional and regional scales. This is due to their geographical location

and to the fact that they are impacted, to a large extent, by similar types of sources. Therefore, selecting any of these cities would be an adequate choice as a representative urban environment of this area. Certainly we will be able to identify other groups of cities for the rest of the U.S. that are impacted by similar types of sources and meteorology; however, because of the limited or non-existent data for other cities, grouping will be more challenging. Final decisions on cities to be investigated will be made based on additional considerations, such as the concurrence of epidemiological, long-term exposure or toxicological studies and the availability of human resources to operate the sites at the specific city.

From the exposure assessment point of view, it is important to mention that activity patterns and home characteristics, which are important determinants of exposure, can vary within these subregions. For example, important differences in both activity patterns and home characteristics exist between Boston and Washington, although these cities experience similar types of fine particles. For this reason, it would be desirable to include at least three types of environments: a) one with harsh winters and mild summers (e.g., New York, Boston, and Chicago); b) one with mild winters and hot summers (e.g., Atlanta, Washington, and Phoenix); and c) one with mild winters and summers (e.g., Seattle, San Francisco, and Los Angeles).

Frequency and duration of measurements

Diurnal, as well as day-to-day, variability in composition and concentration of fine particles is more pronounced than the spatial variability across a metropolitan area. Similarly, inter- and intra-personal variability in the composition and the concentration of fine personal exposures is higher than the spatial one. As such, use of many sites across a metropolitan area will add very little to our understanding of exposures and their relationship to outdoor concentrations. Therefore, daily measurements of 24-hr concentrations should be obtained at a central site. Continuous and semi-continuous monitors for particle mass and composition should be used to obtain information on concentration diurnal profiles.

It is worth mentioning that limited results from previous particle studies conducted in Philadelphia and Boston suggest that there is little year-to-year variability in particle mass and composition. Therefore, as noted earlier, for exposure assessment and receptor modeling studies, it may be advantageous to place a platform and measure pollution in each city for no more than a year. By

conducting a one- or two-year monitoring program per city, it will make it possible to investigate a larger number of cities.

Finally, spatial studies, involving simultaneous measurements at multiple sites may be of little value to the exposure assessment investigations and therefore should be conducted for only a subset of sampling days.

MEASUREMENT NEEDS FOR SOURCE-RECEPTOR RELATIONSHIPS

Prepared by Pradeep Saxena and Glen Cass

Implementation of the NAAQS for fine particulate matter requires that control plans be drafted on the basis of an adequate understanding of how emissions of particles and gaseous precursors lead to outdoor aerosol concentrations at community air monitoring sites. In this chapter, we propose a conceptual theme for a program of atmospheric measurements designed to support determination of source/receptor relationships based on the following important considerations:

- One must think in terms of urban or regional experiments rather than individual observatories (e.g., super sites) in isolation. The purposes for which atmospheric measurements are needed include construction of data sets that will be used as the basis for confirming that source-oriented air quality models are working adequately. These models operate over large geographic areas (e.g., the entire eastern United States or all of southern or central California). In an experiment designed for use in evaluating such models, one or more super sites will serve as the central observatories for gathering the most intensive measurements; observations at perhaps a larger number of other satellite observatories (e.g., sites in the speciation network) will be essential as well. Prior urban and regional particle experiments such as the Southern California Air Quality Study (SCAQS) [Lawson, 1990] can serve as the starting point for designing such an experiment.
- Since the source-receptor relationships that determine fine particle concentrations and regional haze have a fundamentally similar basis, we propose that the experiments be designed to permit both particle concentrations and regional visibility relationships to be studied simultaneously. For instance, to the extent feasible, special experiments in National Parks ought to be synchronized with the NAAQS-based fine particle experiments in urban and rural areas. Doing so will provide the modelers concurrent observations at more locations than they would have otherwise. Similarly, the experiments should be coordinated with ozone formation or continental radiation-balance experiments. In the same vein, coordination with other public sector and private sector efforts can be used to overcome resource-driven compromises in experimental design.

- Although the NAAQS for fine particulate matter addresses only fine particle mass concentration, we need to know the chemical composition of the total aerosol (i.e., both particles and gas-phase components) to confirm the accuracy of models that attribute the concentrations at specific receptors to emissions from specific sources, as well as to provide the information required by health and exposure scientists. For these reasons, our charter is to address the source-receptor relationships for not only fine PM mass, but for fine particle physical and chemical components such as ultrafines, organics and acidity as well.
- The annual average fine particle air quality standard may be the limiting standard for many areas; however, the methods for relating emissions to atmospheric fine particle concentrations (e.g., first-principle emission-based simulation models) that are available today are best suited to episodic applications. This mismatch will have to be dealt with in the future on the basis of prior experience (e.g., by the episode aggregation methods used in the National Acid Precipitation Assessment Program, NAPAP) or by new engineering and computational innovations that produce models that can run in time series for a year. At this time, a safe experimental design will involve a) year-round measurements such that the models can be applied at a basic level when and where needed and b) more intensive episodic measurement programs designed to test the models against a few data sets that are sufficiently well defined that it will be difficult for the models to produce seemingly the right answer for the wrong reasons without that defect being detected and corrected.
- The first generation of comprehensive, first-principle fine particle models have been published in the peer-reviewed literature largely during the last two years. Some of these models have undergone limited evaluation, most frequently for areas in California (e.g., against data from the Southern California Air Quality Study). Given the community's experience with ozone modeling during the last two decades, we expect that the first-generation particle models will undergo an evolution and maturing over the next decade, which will crucially depend upon the availability of observational data needed for testing them. Therefore, it is particularly important to design experiments both inside and outside of California with a vision of their long-term value for diagnosing and improving particle models. Moreover, advanced particle models subsume the essential physics and chemistry of ozone and deposition models; therefore, the model evaluation data sets have to address photochemical and cloud chemistry components.

- Recent studies off the east coast of the U.S. show that the composition of particles aloft can be different from that at the ground level. Therefore, ground-level measurements alone are insufficient to characterize the boundary layer: models that seek to calculate source-receptor relationships should be tested against 3-D measurements of atmospheric transport and concentrations. This can be accomplished via synchronized aircraft and ground-level measurements.
- In addition to the mechanistic source-oriented models discussed above that determine source/receptor relationships by simulating the physical processes in the atmosphere, there are additional models that perform much the same service via chemical tracer techniques. Inorganic, as well as organic, tracers can be used to quantify the contributions of specific sources to concentrations at specific receptors. Using mechanistic models in tandem with these observation-based techniques is the most reliable strategy for source attribution.

Major science questions

Reliable answers to policy-relevant questions derived from model simulations become available only after scientific investigation has produced the tools that are necessary for decision analysis. The scientific investigation itself begins with a conceptual model followed by quantitative investigation to measure important meteorological and chemical conversion processes, followed by tests of the conceptual model to convince ourselves that we are getting the right results for the right reasons. Such investigations can often lead to paradigm shifts in which we find that our prior concepts are wrong. As we proceed through this process, measurements will be needed to help address the following scientific questions:

- What is the concentration and size distribution of fine particles and their important components (inorganic ions, elements including metals, total extractable acidity, organics, soot, and ultrafines) at the receptors of interest (e.g., an urban nonattainment area)? What is the spatial and temporal variability in these concentrations? What are the errors associated with these concentrations?

- How accurately can air quality models translate data on source emissions of gases and primary particles into predicted air pollutant properties at receptor air monitoring sites?
- What are the spatial scales over which emissions sources influence air quality to a significant degree? For instance, for a specific episode or an annual average period, can 95% of the concentration of each important component be attributed to emission sources located within 50 km or 100 km or 500 km of the receptor site? What are the error bars on these judgments?
- Can we meaningfully attribute the total concentration of a specific component (e.g., sulfates or carbon particles) to a number of specific sources (e.g., 20% of sulfate or particulate carbon is attributable to source X, 30% to source Y and so on)? Or is it more appropriate to think in terms of general region-wide changes in emissions versus the changes in concentrations? What are the error bars associated with these judgments?
- Are there any substantial nonlinearities or inter-component couplings in the system? For instance, will the reductions in particulate sulfate concentrations lead to an increase in particulate nitrate concentrations? Or will the reductions in VOC emissions lead to a reduction in ozone but also to an increase in particle-phase organics? What are the error bars on these judgments?
- Can the models be demonstrated to track those changes in emissions that have occurred historically (e.g., the outcome of Title IV in the eastern U.S.; emissions changes in Los Angeles)?
- Can the models simultaneously address source/receptor relationships for both ozone, fine particles, and atmospheric light extinction, as the control programs for these classes of pollutant problems are directed at many of the same sources?

Experiment design: observables, frequency, duration, and location

We propose a multi-year (e.g., 2-3 year) experiment involving a basic set of year-long measurements supplemented by 2 to 6, 1-month long intensive sampling periods distributed during various seasons and meteorological regimes. The intensive experiments will involve observations of more variables at more locations and at higher time resolution than during the rest of the experiment. These

measurements will be used for diagnostic evaluation of those air quality models that are based on first-principles and for their subsequent application to specific episodes to reveal source/receptor relationships. Chemical tracer-based models will utilize data from all ongoing periods of observation (intensives and nonintensives).

As mentioned before, large regional spatial coverage can be obtained by establishing heavily instrumented super sites surrounded by more lightly instrumented satellite sites. The complex central observatories could consist of a combination of EPA-operated super sites and observatories operated by others (e.g., DOE, NPS, universities, private sector). Similarly, satellite sites could be formed from a combination of EPA's speciation network sites, Interagency Monitoring of Protected Visual Environments (IMPROVE) sites, and observatories operated by other parties. Based upon our prior experience, it will be beneficial to plan as many redundant measurements as possible to provide a test of data quality and to ensure availability of data in case of instrument failure or in case inadequacies in an analytical method are later discovered.

The instrumentation used during field experiments (Table 1) will include devices for measuring the following:

- chemical as well as meteorological observables
- surface as well as aloft air
- gas-phase as well as particle-phase components comprising important reactants, products and source tracers, with attention to measurement of both particle size as well as particle chemical composition.

We have included observables that are specifically suitable for testing observation-based models. For instance, in observation-based models, CO can be used as a tracer for urban emissions and SO₂ for power plant emissions. Similarly, data on individual organic compounds have been used for source apportionment by chemical tracer techniques in California and Denver.

Our expectation is that a combination of filter-based and in situ technology will be used in these experiments. For instance, 24-hr average concentrations of ions, organic and elemental carbon, and trace elements can be derived using filter samplers with denuders and back-up filters as necessary to capture any volatilization of particles in order to obtain data over long periods of time that are needed to support models for annual average concentrations. The same or similar samplers can be operated over consecutive 4-hr periods as in the SCAQS experiments to obtain time series data on particle

dynamics during intensive air quality experiments. On the other hand, 1-hr average or nearly continuous concentration measurements can be made for some species using new or proven technologies including aerosol time of flight mass spectrometry. Some overlap between 24-hr, 4-hr, and 1-hour measurements will be necessary to ensure comparability of the data.

The experiment will include urban and nonurban locations. The exact location of the sites and their number will depend upon the region to be studied. Los Angeles should be studied as a region by itself. The same can be said of the Denver area or central California, both of which exhibit noticeable fine particle air quality problems. A study domain in the eastern United States will be much larger and should cover many urban areas. For the east, we propose one central site in each of the following five general areas:

1. Metropolitan New York/New Jersey
2. The Ohio River Valley (e.g., Cincinnati)
3. The Great Lakes area (e.g., Chicago)
4. Metropolitan southeast (e.g., Atlanta)
5. The Gulf Coast (e.g., New Orleans, Houston)

These could be supplemented with experiments conducted by other NARSTO organizations (e.g., DOE's Pittsburgh super site). In addition to the urban central sites, six to eight times as many satellite sites should be identified that fill in the areas between super sites and that will include both urban and rural locations.

We suggest two strategies for keeping the cost under control:

1. Utilize existing or planned stations where some of the observables are already (or planned to be) measured (e.g., PAMS sites, upper-air meteorological stations, and speciation sites) so that this experiment bears only the incremental cost of adding some observables.
2. Instead of measuring concentrations throughout the month-long intensives, do so only for a few sets of days within each intensive. This strategy has been used in SCAQS and other studies.

Table 1. Aerosol & Meteorological Measurements Needed for PM_{2.5} Source Attribution

Observable	Central Observatories (Supersite) ¹		Satellite Observatories ¹		Aircraft ¹
	Sampling Frequency	Sampling Duration	Sampling Frequency	Sampling Duration	Intensives Only

I. Surface Aerosol Composition

Gases

CO	C	Y	C	Y	X
VOC Comp	4/24-hr	I/Y	4-hr	I	X
NO, NO ₂ , NO _y , PAN	C	Y	C	Y	X
O ₃	C	Y	C	Y	X
H ₂ O ₂	C	I	--	--	X
SO ₂	C	Y	C	Y	X
OH & NO ₃	--	--	--	--	X

Multi-phase (Gas & PM_{2.5}) Components

NH ₃ & NH ₄ ⁺	C, 4/24-hr	I/Y	4/24-hr	I/Y	X
HNO ₃ & NO ₃ ⁻	C, 4/24-hr	I/Y	4/24-hr	I/Y	X
Labile Organics	24-hr	Y	--	--	--
HCl & Cl ⁻	24-hr	I	--	--	--
Particle H ₂ O	1-hr	I	--	--	--

Fine Particle Components

Total Mass	C, 4/24-hr	I/Y	4/24-hr	I/Y	X
Sulfate	C, 4/24-hr	I/Y	4/24-hr	I/Y	X
Acidity	24-hr	Y	24-hr	Y	--
Total Organic Carbon	C, 4/24-hr	I/Y	4/24-hr	I/Y	X
Elemental Carbon	C, 4/24-hr	I/Y	4/24-hr	I/Y	X
Organic Comp (including source tracers)	24-hr	Y	24-hr	I	--
Trace Elements	4/24-hr	I/Y	4/24-hr	I/Y	X
H ₂ O-Sol. trans. metals	24-hr	Y	24-hr	Y	--
Particle comp. by size (impactors)	4-hr	X			

Fine Particle Physical & Optical Properties

Particle Size & Number Dist (nm to μm)	C	Y	--	--	X
Light Scattering	C	Y	--	--	X
Light Absorp.	C	Y	--	--	X

Table1 (continued)

Observable	Central Observatory (Super sites) ¹		Satellite Observatories ¹		Aircraft ¹
	Sampling Frequency	Sampling Duration	Sampling Frequency	Sampling Duration	Intensives Only

II. Surface Meteorology

Temperature	C	Y	C	Y	X
Relative Humidity	C	Y	C	Y	X
Wind Speed & Direction	C	Y	C	Y	--

III. Cloud and Fogs

Total Water Content	--	--	--	--	X
Ionic Composition	--	--	--	--	X

V. Boundary-layer Meteorology

Temperature (z)	C	Y	C	I	--
Wind Speed & Direction (z)	C	Y	C	I	--
Vertical Velocity (z)	C	Y	C	I	--
Mixing Depth	C	Y	C	I	--
Relative Humidity	C	Y	C	I	--

VI. Surface Deposition Fluxes	24-hr	Y	24-hr	I	--
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1: C: Continuous (time resolution of ≤ 1 -hr)

Y: Year-long (for the entire duration of the experiment)

I: Intensives only

X: Measure at a frequency and for a duration that are optimal from the standpoint of cost, technological and logistical considerations

04/24-hr I/YÓ means 4-hr resolution during the intensives and 24-hr resolution during the rest of the experiment. 0C, 4/24-hr I/YÓ means continuous & 4-hr resolution during the intensives and continuous & 24-hr resolution during the rest of the experiment.

INTRODUCING ACCOUNTABILITY IN THE MANAGEMENT OF PM_{2.5} AIR QUALITY¹

Prepared by Ken Demerjian

Accountability defines the process and components needed to identify pathways toward attainment of selected environmental goals/standards (in this case the PM_{2.5} NAAQS). Accountability includes: a) demonstrating progress in attaining specified goals/standards, b) quantifying the effectiveness of the management approaches applied to achieving specified goals/standards, and c) the organizational requirements (authority and responsibility) to oversee the process.

The focus of this discussion is on the process and components of an accountable management system and not on the organizational aspects of its implementation.

Given the considerable costs to be expended annually to address the new PM_{2.5} environmental regulation, the public has the right to ask the scientific and policy communities to evaluate the effectiveness of implemented environmental controls both in terms of meeting air quality standards and anticipated improvements in environmental health. As with any management system, it is reasonable to expect that analytical measures be in place to demonstrate the progress, success and failure of the air quality management system. The identification of an accountable PM_{2.5} management approach is essential to a credible pollution mitigation program. This is particularly true for PM_{2.5} given the complexities of the physical and chemical processes involved in its production and distribution in the atmosphere

Basic components of an accountable PM_{2.5} air quality management system

Irrespective of the approach used to design the emission control strategies adopted to meet the PM_{2.5} standard, a framework for assessing progress and demonstrating the success or failure of prescribed actions should be an essential feature of the management approach. The three principal steps required for implementing an accountable PM_{2.5} air quality management process are:

- ¥ Verify that implemented PM_{2.5} primary and precursor emission controls are performing according to specifications.
- ¥ Verify that PM_{2.5} air quality has responded to the emission changes achieved as expected.
- ¥ Verify that the response of identified public health and welfare receptors agree with expectations given the observed changes in PM_{2.5} air quality.

Typically these steps occur in serial order and are increasingly more difficult to perform as one proceeds through the list.

¹ Adapted from Demerjian, et al., 1995 and Chapter V of the NARSTO Ozone Assessment, 1998 (in review)

- Step 1 involves the testing and evaluation of the PM_{2.5} primary and precursor emission controls implemented and verifying that these control measures do in fact comply with specifications and established requirements.
- Step 2 demonstrates that PM_{2.5} air quality has responded in the expected way to the emission reduction documented in step 1. This would include monitoring ambient air changes in PM_{2.5} mass and chemical composition, changes in their precursor concentrations as well as changes in the wet and dry depositional loading of select pollutant species with time.
- Step 3 demonstrates that the observed changes in PM_{2.5} air quality documented in step 2 have resulted in an expected and quantitative benefit to public health and welfare. Because of confounding factors, this last step is the most difficult to demonstrate and it may take many years to establish credible data for verification.

Introducing accountability in the management of PM_{2.5} air quality clearly depends on the successful design and deployment of measurement networks capable of providing good quality spatial and temporal data on PM_{2.5} mass, chemical composition and its relevant precursors. It also depends on the development of a substantially more detailed, long-term surveillance system for indicators of human and ecosystem health than is currently in place.

The PM_{2.5} attainment demonstration process: A new approach

The current approach to air quality management stipulates control programs based on "engineering estimates" that are intended to achieve needed reductions in primary and/or precursor emissions. These control programs are typically projected through model simulations. As currently configured, the management process does not require verification that implemented emission controls have achieved expected changes in precursor concentrations in the atmosphere. There is a significant need to develop analytical procedures, which utilize air quality measurements to track emission changes in the environment and demonstrate the attainment of emission reduction goals as specified in air quality management plans. The role of air quality modeling systems in the implementation of the PM_{2.5} management approach has yet to be defined, but if it follows a track similar to that of the ozone attainment demonstration process, it could benefit significantly from acknowledged limitations in that process [Demerjian, et al., 1995]. The development of control strategies for the attainment of the PM_{2.5} standards will likely proceed through the combined use of diagnostic and prognostic modeling systems. If the scientific tools to be developed and applied in the PM_{2.5} attainment demonstration process are to be fully accountable they should include the following essential features:

- Demonstrate through the direct measurement of PM_{2.5} primary and precursor source emissions that specific emission control programs are meeting their objectives and maintaining their expected effectiveness in time, and that changes in emissions track inventory estimates and projected reductions as expected.
- Demonstrate through air quality measurements of PM_{2.5} primary and precursor species that precursor concentrations in the airshed have responded as expected to changes in precursor emissions.

- Demonstrate through air quality measurement that $PM_{2.5}$ mass and chemical composition in the airshed has responded as expected to the changes in precursor concentrations as a result of implemented controls.
- Demonstrate through improvements in health-related measures and ecosystem responses that changes in $PM_{2.5}$ air quality have achieved expected health and welfare benefits.
- Help provide appropriate feedback mechanisms through evaluation, formulation, implementation of the scientific tools to consider appropriate alternate abatement strategies. If demonstration fails at any point in the above sequence, re-initialize the demonstration process.

Implications for the measurement system

A well structured air quality monitoring system plays a key role in the implementation of an accountable $PM_{2.5}$ air quality management system. It should be our "watchful eye", providing an early indication of success or failure, while indicating where mid-course corrections may be warranted. The basic components of such a system, as described above, provide specific guidance for the measurements being discussed.

In order to verify that $PM_{2.5}$ air quality has responded to emission controls in an expected way the system must be capable of quantifying $PM_{2.5}$ mass (including chemical speciation of primary and secondary particulate mass) and concentrations of PM precursors. Precursor species of interest would include nitrogen oxides, volatile organic compounds, sulfur dioxide and ammonia. Speciation of the $PM_{2.5}$ mass will provide additional information on the effect of emission reduction strategies. The emissions of many of these precursor compounds are being reduced in response to other environmental concerns (e.g., acid rain and ozone) and collateral benefits may accrue to reduce ambient PM concentrations.

In terms of health, "accountability" requires that first, the sites be adequately representative of the larger U.S. population, so that effects seen around the sites can be extrapolated to the Nation, and that there be a focussed effort to build both routine and special purpose health status monitoring capabilities around each site. These would include routine collection of detailed health indicators, identification of representative study cohorts of individuals to be followed over 20 or more years, and identification of key health quality indicators.

The time scales of the changes in emissions and the health indicators are such that multi-hour (1-12) averages are adequate to evaluate the performance of the $PM_{2.5}$ air quality management plan. However, both the quantification of air quality benefits and the demonstration of improvements in community health indicators requires that measurements be conducted over an extended period of time (several years to a decade). Year-to-year variation in meteorology can mask improvements in air quality that result from changes in emissions. Measurements must be made over a long enough period of time that the effects of the meteorological variability can be dealt with in a statistically robust fashion. In a similar fashion, changing lifestyle and demographic factors that confound the analysis of health effects data require larger data sets for successful analysis.

DEVELOPMENT AND EVALUATION OF PM MEASUREMENT METHODS

Prepared by Susanne Hering, Pradeep Saxena and Jim Meagher

The measurement of the concentration, chemical composition, and physical characteristics of ambient aerosols is a daunting task. Unlike ozone or carbon monoxide, ambient PM is not a single chemical constituent. Instead, it refers to a phase, either liquid or solid, that may be in a delicate equilibrium with the surrounding atmosphere and that consists of hundreds of compounds. Slight alterations in temperature and/or humidity that may occur during sampling can significantly alter the characteristics, composition, and mass of the aerosol material.

The difficulty and expense of PM measurements has significantly impaired our ability to understand the processes that control the formation and distribution of PM in the atmosphere, in our homes, factories and offices and in our personal breathing zones. In order to remedy this situation we need to foster the development of new methods and technologies and provide the means to evaluate existing and emerging methodologies.

Development, evaluation and intercomparison of methods and technologies should be an important part of any development program. Reference standards (e.g., material of known composition that can be used to test the accuracy and precision of an instrument) are needed that are robust, easy to use, and accurately mimic the characteristics and composition of atmospheric aerosols. Carefully planned and executed PM measurement intercomparisons that utilize common standards in the laboratory and side-by-side sampling in the atmosphere are needed to provide comparative data for different techniques that measure the same aerosol parameters. As new techniques emerge there will be a growing need to test their accuracy and precision as well as their relative performance for differing environments (i.e., ambient air, indoor air, and personal exposure).

In the near term (1-3 years), efforts to improve and expand PM measurements should focus on three areas, which are described below.

Evaluation of the Federal Reference Method (FRM)

The EPA's guidance to the States requires that, by the end of 1999, approximately 850 regulatory monitoring sites be established throughout the U.S. for the measurement of $PM_{2.5}$ mass. Data from the FRM samplers located at these sites will be used make attainment/nonattainment designations with respect to the $PM_{2.5}$ National Ambient Air Quality Standard (NAAQS). Additional information on the national monitoring network is provided in Appendix A.

The $PM_{2.5}$ FRM has been extensively tested by the EPA and, like all such methods, it has strengths and weaknesses. The single-filter impactor technology for measuring $PM_{2.5}$ mass has been around for some time and has been engineered to reliably sample the nonvolatile component of fine particles. The most pressing weakness, to paraphrase and quote from the NRC report, is the potential for loss of volatile material (e.g., nitrate, chloride, some organics) that can transition from particle to gas phase (or vice versa) during or after sampling. The loss of volatile material is likely to depend upon location and season. Some experiments have already shown the magnitude of this loss for locations in California; data from tests in other parts of the country are likely to become available by the end of

this year. If these losses turn out to be substantial, then using the FRM would amount to quantifying only a fraction of the outdoor concentration and that fraction would vary geographically and seasonally. Alternative technologies that overcome some of these biases could be developed and, over time, then compared, evaluated and adopted.

Development/evaluation of PM methods for ambient air

As mentioned in other parts of this report, the super site network alone cannot meet the needs of epidemiologists, exposure scientists and atmospheric modelers: therefore, super sites have to be an integral part of a broader experiment. In this connection, EPA, with assistance from the States and others, will establish a national network of 300 routine chemical speciation sites (Appendix A). The type of chemical speciation network that has been proposed will be extremely valuable and conceptually a complement to the super site network. Therefore, it is fruitful to consider design enhancements to the speciation network in order to render the super site and speciation networks as useful adjuncts to each other. With respect to the current proposal for the speciation network, we recommend that the following enhancements be explored:

- Time resolution**, which is an important issue for understanding health exposures and particle origins. The 24-hr integral filter techniques most commonly used in aerosol studies do not provide the time resolution needed to evaluate source attribution simulations including some of the observation-based methods. For many observables we need to characterize ambient aerosols on shorter time scales (< 1 hr). Needed are automated methods for the physical and chemical characteristics of particles that may be relevant to health (personal exposure), to the understanding of particle sources and process dynamics, and to evaluating air quality changes that may result from controls.

Recent advances include automated methods for characterizing the chemical composition of short-term (e.g., 15-minutes) particle samples as well as single particles in situ. In the near future, these technologies may become attractive due to their greater accuracy and lower operating costs. For some observables (e.g., sulfate) techniques for measuring particle phase concentration at sub-hour resolution are already commercially available; for others, research grade methods have shown promise. Some investment is needed for testing these methods in the laboratory to characterize and improve their accuracy, reliability and fieldworthiness. Moreover, providing platforms and support for the field testing and intercomparison of new methods (including comparisons with more traditional, albeit labor intensive, methods) is critical to their development and practical and extensive application as required by EPA and the states to protect public health.

- Data immediacy**, which is important - even necessary - if ultimately the public is to be warned of episodes. Fast response measurements and rapid data turnaround are also important during intensive field campaigns, where continuous measurements can guide the deployment of resource-intensive measurement systems, such as instrumented aircraft, and sample collection for detailed chemical analysis.

- Organic aerosol sampling and composition**. Due to the volatility of organics, accurate sampling of organics is a notoriously difficult problem. Particle samples can suffer positive and negative artifacts due to adsorption and evaporation of volatile organics respectively. Promising technologies such as denuders, particle concentrators and post-filter media for capturing semi-volatile organics need to be tested for widespread deployment. Prototype samplers could be located at PAMS sites to take advantage of the suite of measurements that are available at these sites, in particular the volatile organic compound (VOC) analyses.

The speciation of the organic fraction of ambient aerosols is a critical need for determining the causative agent for health damage as well as for source attribution. So far, typically the composition of only 10% of the total organic carbon has been characterized; even such data on the molecular composition are rare. Data are particularly sparse for the polar organics that are more water soluble. The characterization of organics in aerosols is confounded by chemical transformations that occur during collection and by rapid exchange between the gas and solid phases. A number of promising ideas and techniques exist that would benefit from laboratory and field evaluation and intercomparison.

Development/evaluation of PM methods for personal exposure

PM research requires that we be able to characterize air in three regimes: outdoor air or the ambient atmosphere, indoor air in our houses, factories and offices, and our personal breathing zone. Most of the PM measurement methods in use today were developed to characterize the ambient atmosphere. Although sampling the indoor environment presents some unique challenges, many of the same techniques can be applied, with minor modifications, to that environment. Characterizing the personal breathing zone, however, presents the greatest challenge.

Methods currently in use to monitor personal PM exposure rely primarily on the collection of PM samples by impaction on filters. For example, *Marple et al., 1987* developed a personal PM₁₀ sampler that was used in the Total Human Environmental Exposure Study (THEES) [*Lioy et al., 1990*]. The same sampler was also used in the 1990 Riverside CA Particle Total Exposure Assessment Methodology (PTEAM) [*Thomas et al., 1993*] study. This sampler is bulky, heavy, and sample collection is limited by battery power. The method gives an integrated sample over a time period usually lasting 24 hours. The sampling time is selected for practical reasons having to do with battery life for the personal pump and the need to collect enough mass to analyze. Less than 24 hours and one does not collect enough sample to weigh or analyze for chemical constituents; more than 24 hours and the pump battery fails.

It is very difficult to construct an accurate personal exposure model using filter data with long (12-24-hr) averaging times. New technologies are needed to provide finer time resolution estimates of personal exposure. These systems need to be relatively inexpensive if enough individuals are to be monitored to provide a database with sufficient statistical power to describe exposure that is representative of the general population.

Implications for the measurement program

What is to be measured?

The FRM should be collocated with samplers that minimize or eliminate nitrate and organic artifacts. Full particle size, number, and perhaps surface area distributions should be obtained using multiple techniques to cover the entire range from nanoparticles to 10 µm particles. Particle samples for chemical analysis should be collected in additional size ranges for chemical composition (i.e. ultrafines, if possible). One should also run samplers that tie to other networks that may be used in the future (U.S., Canadian, and Mexican) - including visibility monitors to obtain correlations. Meteorological parameters that influence collection efficiency (e.g. temperature, dew point) should also be measured.

Where are the measurements to be made?

PM_{2.5} super sites should be situated (possibly co-located with operational PM_{2.5} network monitoring sites) such that they are available to benchmark and evaluate operational measurement systems. Intensive study areas should be situated at both urban and rural/regional locations.

In the larger picture, the greatest benefit will be realized by placing intensive monitoring sites surrounded by high-level speciation sites, all designed to support one or more sets of data analysis. These platforms could easily provide simultaneous data for: a) epidemiological studies b) methods comparisons and c) aerosol characterization.

When will the measurements be made?

A combination of short-term intensive measurements and longer-term measurements is needed to evaluate the newer short time response instruments and the FRM respectively. It is critical that the measurements be made over a sufficiently long period of time that the instruments under study are exposed to a considerable range of aerosol concentration and composition.

TOWARD AN INTEGRATED RESEARCH PROGRAM: ISSUES FOR THE WORKSHOP

The five preceding sections contain initial thoughts from the Workshop Steering Committee on the design of a research measurement program for PM related research. The suggestions offered are naturally influenced by differing perspectives and differing objectives appropriate to each of the areas of interest and issues addressed. Each of these perspectives and objectives places different demands on the program design in terms of the species measured, measurement frequency and location. Our goal is to review in depth, during the Workshop, each of these perspectives and objectives, and then to synthesize them into an integrated research measurement program that can be implemented jointly by EPA, the state and local environmental agencies, and the health effects, exposure assessment, and atmospheric sciences research communities.

We posed in the Introduction a series of questions to help provide some focus and structure for the suggested program design elements. A sampling of the responses to these questions is repeated below to provide a sense of the program scope required to address the needs identified by these diverse science communities.

What are the major science questions/hypotheses?

The current hypotheses regarding those characteristics of PM that may be responsible for the observed adverse health effects are listed in the health effects section. Although the list may be incomplete and uncertain it provides an important linkage between the health effects and measurement communities. In most cases the hypotheses presented point to a particular parameter or parameters whose measurement will facilitate the testing of the hypothesis.

What is to be measured?

For the purposes of personal exposure assessment it is argued that due to the uncertainty regarding the agent(s) responsible for the health effects a wide variety of parameters relating to PM morphology and composition need to be measured.

Where are the measurements to be made?

The focus of many of the health studies is in urban centers with lots of people that adequately represent the larger U.S. population. However, an adequate understanding of the role of transported pollution requires that measurements be made in rural areas, upwind and downwind of population centers.

When will the measurements be made (frequency and duration)?

Three different time scales are evident. Health outcomes occur on time scales that vary from hours to days to years. Therefore the health effects community may require high time resolution as well as daily, seasonal and annual information to test their hypotheses. The dynamic nature of the chemical and physical processes that influence PM formation and transport requires high time resolution (<1 hr) data to reliably establish source/receptor relationships.

Clearly, these questions evoked very different responses and needs from the different interest areas. However, there are several areas of overlap where specific measurements and measurement strategies can serve multiple objectives. A few of these are provided below by way of example and to stimulate additional ideas along this line during the Workshop.

- In several cases there was a need expressed to measure the same, or similar, wide variety of parameters. These included not only measurement of PM characteristics but also gaseous co-pollutants and meteorological parameters. This broad-based approach was driven by the uncertainty over the agent(s) responsible for the health effects and the need to understand the processes responsible for PM formation and distribution in the atmosphere.
- Many of the objectives of a source/receptor study could be met at one or more of the cities where a major epidemiological study is also underway. The measurements taken to elucidate atmospheric processes could enrich the health effects study.
- Several of the measurement programs suggested would benefit enormously from the concept of using mobile sites in a scoping study to identify suitable areas for intensive research. The scoping studies could be used to identify areas having specific contrasts (e.g. similar climate, activity patterns, and ozone levels but different PM characteristics). Such studies would allow sites to be selected that could be used for multiple purposes (i.e. health effect, exposure, source/receptor studies and instrument testing).
- Many of the suggested measurements are already being made, or will soon be made, at sites across Mexico, the U.S. and Canada. It is essential that these measurement sites be incorporated into the program design where feasible. Whenever possible, resources should be used to extend or augment the measurements currently being made at these sites rather than establish new sites. The U.S. PM speciation network (Appendix A) is a good example of such an opportunity. These sites will provide a valuable subset of measurements that can be usefully extended to provide additional measurements. Operating an intensive site in an area with existing PM measurements will provide additional information of spatial and temporal variability that is important in developing personal exposure models and provides opportunities for methods intercomparison.

Next steps toward implementation

Given the wide variety of participants from Federal (both U.S. and international), state and local agencies, and from the several research communities, it will be necessary, following the Workshop, both to synthesize these elements into an integrated research management program and to establish an ongoing mechanism for collaboration and coordination among participants to design and implement the program. The exact form of such a mechanism needs to be developed to take advantage of existing structures (e.g., NARSTO) while ensuring that the broader research community is actively engaged.

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APPENDIX A

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OVERVIEW OF NATIONAL PM_{2.5} MONITORING NETWORKS

The current planned scope of the national PM_{2.5} network consists of three major components: Mass monitoring, routine chemical speciation and special study areas termed “super sites”. In very broad terms, the network as a whole supports three principal regulatory objectives: 1) Determining nationwide compliance with the NAAQS, 2) State Implementation Plan (SIP) development (e.g., source attribution analysis and air quality model evaluation, and 3) tracking trends and progress of emissions reduction strategies. EPA recognizes that, with care in design and execution, components of this program can also provide significant support for priority research needs. The following brief description is intended to provide background for understanding the context and relationship among these components and between them and EPA’s research program. The attached table outlines for each category below a synopsis of the budgeted number, major purposes, and potential flexibility for integration with PM research programs.

Mass monitoring (1100)

1. Core mass monitoring (850). Approximately 850 NAMS/SLAMS sites, required according to EPA guidance to the States, will be dedicated to mass monitoring. A breakdown of these 850¹ sites includes 750 required for NAAQS compliance and 100 sites for characterizing background and transport. The regulation requires a continuous sampler to be collocated with an FRM/FEM at the 52 largest cities (greater than 1,000,000 population).
2. Mass samplers for spatial averaging and special purpose monitoring (SPM)(200). Roughly 200 additional sites to accommodate spatial averaging² and special purpose monitoring needs are expected to be deployed. The SPM sites are those established to identify unique source location or communities, and are not required to be compared to the NAAQS if operating less than 2 years (or a sampler without FRM/FEM designation).
3. Continuous monitoring (50). In addition to the required collocated 52 continuous monitors, plans include deployment of an additional 50 continuous samplers. Collectively, at least 100 continuous samplers will be deployed, and probably more, since the States can elect to purchase and operate continuous samplers for sites designated as special purpose monitoring.

Principal objectives for mass monitoring:

- (a) FRM/FEM samplers and NAMS/SLAMS. The primary objective for mass monitoring, especially the designated NAMS/SLAMS¹ sites, is for comparison to the PM_{2.5} NAAQS. In addition, 100

¹ EPA network guidance (40CFR58) requires 850 NAMS/SLAMS sites; however, 100 of those sites are to be designated as background or transport sites (2 per State) which can use the IMPROVE sampler, which is not designated as an FRM/FEM and therefore would not be used for NAAQS comparisons.

² The annual PM_{2.5} standard is specified as reflecting an area-wide distribution or spatial average of a representative single monitor or the average of multiple monitors. States have requested additional monitors to provide for spatial averaging.

NAMS/SLAMS will serve as background and transport sites, integrated with other efforts such as the Interagency Monitoring of Protected Visual Environments (IMPROVE) program, to characterize regional transport and background concentrations.

- (b) Continuous samplers. Continuously operating samplers will provide a real time estimate of $PM_{2.5}$ levels and allow for input into public information displays (similar to current ozone mapping efforts that reach local weather forecast venues) as well as the Pollutant Standards Index (PSI). Other objectives for continuous samplers include developing statistical relationships with FRM/FEM's to serve as potential surrogates for compliance indicators, and characterizing diurnal patterns of exposure and emissions.
- (c) Special Purpose Monitors (SPM's). The SPM samplers are intended to provide flexibility for State and local agencies to investigate areas that may have exceedances without the repercussion of regulatory requirements associated with NAAQS violations. The purpose of SPM's is to encourage monitoring where it might otherwise be discouraged due to fear of associated regulatory requirements. The SPM's are expected to be located in unique or rural communities subject to localized sources, or enhance the regional/background/transport network to better characterize multiple spatial scale interactions. Samplers for SPM purposes can be FRM/FEM that operate less than 2 years, or non- FRM/FEM samplers. Many State and local agencies are expected to operate continuous samplers within the classification of SPM sites.

Routine chemical speciation (300).

The routine chemical speciation program consists of two components: 50 required NAMS, and up to 250 additional sites (EPA's contribution to the IMPROVE program technically is similar to the routine speciation program but addressed separately due to budget considerations). The major purpose of these sites is to assess long-term trends in major $PM_{2.5}$ components, as well as to provide useful information for source apportionment, evaluating current and future control programs, and health risk assessments.

1. NAMS (50). The regulation requires 50 speciation sites across the country, located mostly in urban areas (e.g., all PAMS cities will have a speciation site). These 50 sites will be designated as NAMS and will follow sampling and analysis protocols similar to the existing IMPROVE program. Filter sampling techniques (teflon, nylon and quartz media) for 24-hour periods will be analyzed for principal mass components: most elements through X-ray Fluorescence; major ions through Ion Chromatography/Colorimetry (nitrates, sulfates, chloride/ammonium); and organic and elemental fractions of carbon through thermal analysis. The sampling methodology and frequency (1-in-6 day or greater) are being evaluated in light of peer review comments. Prescriptive protocols for sampler selection, analytes, and sampling frequency will be adhered to ensuring national consistency across space and time.
2. Other "Routine" Speciation Sites (250). In addition to the NAMS, resources are expected to be available to support up to 250 additional sites. These sites will be less prescriptive than the NAMS and will be subject to a balance among competing needs for national consistency (50 sites are not adequate to characterize the U.S., suggestions for more frequent sampling), and flexibility to address local-specific issues such as wintertime wood smoke or the need to support related scientific studies, which might require more intensive seasonal sampling and analysis. This component of the program does provide true flexibility for State and local agencies. Certain States (e.g., California) have expressed an interest in establishing more advanced methods capable of in-situ, near continuous measurements of principal species. Given the flexibility of this component of the National program, substantial opportunity exists to interact with the health and atmospheric chemistry research

communities. With the exception of the super sites program, however, all of these components are funded by State Grants, which provide hardware and related capital costs, laboratory analyses, and salaries for State and local agencies to operate the network. Consequently, the dialogue must involve EPA, State and local agencies, and the research community.

3. IMPROVE Sites (108). In addition to 30 existing EPA supported sites, 78 new IMPROVE sites are being added, in or near Class I Federal areas (e.g. National Parks and Wilderness), to address the requirements of the forthcoming Regional Haze regulations. These sites conduct speciation sampling similar to the 50 NAMS, but on a 1/3 day sampling interval. These sites are considered as part of the entire PM_{2.5} National network, recognizing that the technical connections (e.g., sources/ambient characterizations, measurement techniques) between PM_{2.5} and visibility require integration. Although funded through State Grant funds, this program is managed by the IMPROVE Steering Committee, and most of the technical work is conducted by Universities and the Federal Land Managers.

Super sites (4-7).

EPA plans to conduct special detailed chemical and physical characterization studies in 4 to 7 areas that reflect a range of characteristic PM_{2.5} source-receptor and health risk situations. The scope and specific details of this program, termed “super sites,” are being developed through substantial input from the scientific community, including the July 22-23rd workshop in RTP. The following discussion outlines EPA’s overall objectives for the program and the relationship to other components.

The major objectives common to all of the “super site” study areas include elucidation and study of source-receptor relationships to enable improved implementation and tracking of strategy effectiveness in the overall PM program, providing a basis for improved health risk assessments, and serving as vehicle for comparing emerging sampling methods with routine techniques to enable a smooth transition to advanced methods. The first two objectives reflect an attempt to increase the temporal, chemical, phase and size fraction resolution of measurements relative to “routine” monitoring programs that typically are limited, for example, to intermediate averaging times (e.g., 24 hrs.) and single size ranges.

To optimize the use of these resources for the scientific priorities identified by the NAS panel, planning for the super site program is being integrated with EPA’s PM research planning. The kinds and extent of equipment and the spatial and temporal extent of monitoring in each study area will be tailored to address one or more additional objectives related to the research program. Specific research needs being evaluated include improved source apportionment methodologies, exposure assessment studies, diagnostic studies to elucidate atmospheric process dynamics associated with the formation, accumulation and removal of PM_{2.5} constituents and other associated (e.g., oxidants) atmospheric species, epidemiology studies, and interactive analyses to support toxicology.

This integrated research/super site area planning is also taking into account the speciation and continuous mass monitoring programs outlined above. Each of the special study areas will be points of focus for these programs as well. The spatial requirements for characterizing the multiple interacting spatial scales (horizontal) can not be addressed by super sites and in this context routine sites can be viewed as satellites for greater spatial detail. The super sites can also provide vertical scale resolution (through optical techniques, elevated platforms, periodic aircraft flights) not expected to be part of routine networks, but nonetheless important for addressing the research and regulatory needs outlined above.

Scientific review of network components

The use of PM_{2.5} mass as an “indicator” for PM standards was recommended by the Clean Air Scientific Advisory Committee (CASAC) at the conclusion of their review of the scientific criteria and standards. Both the Federal Reference Method for measuring PM_{2.5} mass and EPA’s guidance for establishing the mass compliance network were peer reviewed by the Fine Particle Technical Monitoring Subcommittee of CASAC in 1996. The more recent plans for speciation measurements, continuous monitors, and super sites are in partial response to the subcommittee’s recommendations for monitoring beyond 24-hour PM_{2.5} mass. The approach for the required speciation monitoring network was recently reviewed by an expert scientific panel in Seattle. The approach and objectives for the super site program will be the focus of a major two day workshop in July. A workshop planning group, including a number of recognized scientific experts in health, exposure, atmospheric sciences, and monitoring met in May and is continuing to develop materials for the program. In addition to providing periodic updates on this program to the NAS panel, EPA intends to present its approach for integrating the “routine” speciation network with the super site monitoring and research programs for review by the Fine Particle Monitoring Technical Subcommittee of CASAC in the Fall.

List of Acronyms

PM_{2.5} = Particles with an aerodynamic diameter less than or equal to a nominal 2.5 micrometers

PM = Particulate Matter

EPA = Environmental Protection Agency

NAMS = National Air Monitoring Station

SLAMS = State/Local Air Monitoring Station

NAAQS = National Ambient Air Quality Standards

FRM = Federal Reference Methods

FEM = Federal Equivalency Methods

SPM = Special Purpose Monitoring

IMPROVE = Interagency Monitoring of Protected Visual Environments

PSI = Pollutant Standards Index